

AD-A134 736

(12)
AEE 401 C24

Contractor Report ARLCD - CR - 83018

M774 MACHINE CHIP RECYCLING

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Prepared for

Department of the Army
U. S. Army Armament Research and Development Command
Dover, New Jersey 07801

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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER Contractor Report ARLCD-CR-83018	2. GOVT ACCESSION NO. AD-A134 736	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) M774 MACHINE CHIP RECYCLE	5. TYPE OF REPORT & PERIOD COVERED Final Report September 1980-June 1983	
	6. PERFORMING ORG. REPORT NUMBER	
7. AUTHOR(s) Mark J. Walz and Charles E. Latham-Brown Nuclear Metals, Inc. W. Sharpe, Project Engineer, ARRADCOM	8. CONTRACT OR GRANT NUMBER(s) DAAK10-80-C-0246	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Nuclear Metals, Inc. 2229 Main Street Concord, Massachusetts, 01742	10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS	
11. CONTROLLING OFFICE NAME AND ADDRESS U.S. Army Armament Research and Development Command STINFO Division (DRDAR-TSS) Dover, New Jersey 07801	12. REPORT DATE	
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office) ARRADCOM, LCWSL Munition Systems Division (DRDAR-LCU) Dover, New Jersey 07801	13. NUMBER OF PAGES 124 pages	
	15. SECURITY CLASS. (of this report)	
	15a. DECLASSIFICATION/DOWNGRADING SCHEDULE	
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution Unlimited		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Depleted uranium (DU) DU-0.75 Ti alloy M774 penetrators DU-6Nb alloy DU-7Nb alloy		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) The work described in this report details the program and the efforts to develop a method by which machine turnings of DU-0.75 Ti could be recycled. The methods addressed are: conversion of machine chips to UF ₄ ; co-reduction of chips in the UF ₄ /Mg reaction vessels; inductoslag remelting; vacuum induction remelting, and electron beam remelting. Indications are that the Electron-Beam method holds the greatest degree of potential in that it has the capability of		

decreasing impurity levels during the melt cycle. However, EB involves a high capital investment and has some technical uncertainties which could only be answered in large scale production.

Vacuum induction remelting is also a potentially viable recovery method; however, it cannot reduce impurity levels. Indeed, it will contribute to progressively higher carbon levels as generations of material are reclaimed.

A specification for an electron-beam vacuum melting and casting furnace has been included as an appendix.

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Unannounced	<input type="checkbox"/>
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ACKNOWLEDGMENT

The authors would like to acknowledge the guidance and assistance of Carl Edstrom in conducting the electron-beam study at the Rocky Flats Plant of the Rockwell International Corporation.

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INTRODUCTION

Objective of the Program

The objective of the efforts described in this report is to respond to the high machine chip scrap disposal costs and volumes generated with the manufacture of depleted uranium (DU) M774 penetrators. The program was initiated to examine chip recycling alternatives, the end of which would produce material of sufficient quality for recycling back into penetrator production. Recovery of such material would ultimately result in reduced penetrator manufacturing costs and in contract savings to the government. This project was funded under U.S. Army Manufacturing Methods and Technology Program 5796634.

Scope of the Research and Development Program

The scope was originally established to evaluate the feasibility of M774 penetrator machine chip recovery using methods which would lend themselves to a viable production system. This would then result in the elimination of certain techniques from further consideration. As new knowledge was gained, the direction of the efforts were altered to study the most promising alternatives.

BACKGROUND

Depleted uranium machine chips can vary considerably in condition and level of recycling difficulty. Important factors to be considered include:

- Ratio of heavy to fine machining cuts utilized
- Cutting speeds and feeds employed
- Oxidation level affected by:
 - a. Type of machine tool involved
 - b. Amount of water soluble coolants used
 - c. Method of dispensing coolants
 - d. Storage method and time frame between chip generation and recycle processing

An extensive amount of work can be put into these areas to maximize the material quality of machine chips destined for recycling. In our experiments, work concentrated on recovering a chip of good quality that is produced during finish machining of premachined blanks used in M774 penetrator production.

In the future, chip quality may be improved to increase recycle yields by examining those factors which affect chip oxidation levels at the CNC machining lathes.

The Drying Process

Over the past few years, Nuclear Metals, Inc. (NMI) has attempted to directly vacuum induction remelt (VIR) briquetted machine chips. The high surface area to volume ratio of the fine machine chips result in rapid and extensive oxidation of the reactive surface area.

Oxidation is enhanced in moist environments so that wet machine chips degrade rapidly. As such, this then results in the reduction of potential metal yields from recycling and further increases the difficulty of reclaiming usable metal (free of oxide inclusions). Dry chips are further required in order to prevent the sudden violent evolution of steam which can occur should moisture come in contact with either molten metal or salts as were planned in the experiments. The drying method developed uses the following process steps:

1. Spin dry to remove bulk moisture
2. Submerge chips in hot oil (300-400°F) to flash off water
3. Degrease in tricholoroethane to remove oil
4. Air dry to remove tricholoroethane

This procedure allows the transfer of hot chips from the oil to the degreaser without experiencing autoignition of the chips. The oil provides a protective layer on the chips which excludes air and thereby prevents combustion. This has proven to be a safe and effective method of thoroughly drying chips for further processing.*

* The drying method described was developed by NMI as a result of internal development of alternative scrap disposal methods. The process has a patent pending on file at the United States Patent and Trademark Office.

METHODOLOGY

Conversion of Machine Chips to UF₄ (Green Salt)

Conversion of uranium metal to uranium tetrafluoride (UF₄) has been achieved by British researchers using a gaseous fluorination technique. Due to the hazards of the British process, NMI performed hydrofluorination experiments with HF acid and DU chips. The basic technique produced UF₄ at 90% purity; however, the 10% balance proved to be uranyl fluoride (UO₂F₂). This compound interferes with the reduction reaction between UF₄ and magnesium. An approach was proposed to examine ways of inhibiting the generation of the uranyl fluoride byproduct and to increase the UF₄ conversion yield. This method was dropped from further consideration at a mid-effort program review due to funding limitations requiring NMI to narrow the alternatives to be evaluated.

Co-Reduction of Chips in the UF₄/Mg Reaction Vessels

This technique was not a true reduction but a method of melting chips with the excess heat generated in the exothermic green salt reduction reaction. In order to evaluate the merits of this technique, a miniature, one kilogram reduction vessel was used for the experiments.

Table 1 lists the eighteen experimental mixtures which were reacted and evaluated for metal/slag separation and yield. The weight of magnesium added to the blends corresponded to the stoichiometric amount required to reduce the UF₄, plus 3% excess.

Trial runs one through four were baseline material runs that did not have chips added to the UF₄/Mg blends. Material quality obtained was good to excellent.

Table 1. Co-Reduction trial runs

Total charge wt. grm.	Run no.	Wt UF ₄	Wt mg.	Wt chips (g)	Wt Li (g)	Initiation temp(°F)	Ending temp(°F)	Yield (%)	Metal quality
1400	1	1207	193	-	-	1071	1208	90.20	Good
1400	2	1207	193	-	-	1010	1184	89.90	Excellent
1400	3	1207	193	-	-	1039	1308	88.79	Good
1400	4	1207	193	-	-	1003	1288	88.06	Good
1342.49	5	1121	179	42 gms	-	1047	1274	94.37	Fair
1342.49	6	1121	179	42 gms	-	1021	1272	73.30	Fair
1342.49	7	1121	179	5 gms	-	Did not React	-	-	-
1337.28	8	1121	174	42 gms	-	1017	1298	79.24	Excellent
1337.28	9	1121	174	42 gms	-	1006	1276	89.80	Excellent
1337.28	10	1121	174	42 gms	-	1040	1316	94.33	Excellent
1345.78	11	1121	174	51 gms	-	1027	1289	77.33	Fair
1345.78	12	1121	174	51 gms	-	1053	1310	78.89	Good
1345.78	13	1121	174	51 gms	-	1022	1294	75.00	Fair
1354.28	14	1121	174	59 gms	-	1017	1322	80.68	Fair
1354.28	15	1121	174	59 gms	-	1003	1288	84.29	Good
1354.28	16	1121	174	59 gms	-	1029	1322	85.18	Good
1361.05	17	1121	174	59 gms	6.8	842	1182	60.45	Poor
1361.05	18	1121	174	59 gms	6.8	940	1246	42.65	Poor

Trial runs 5 through 16 attempted to melt various quantities of chips with the heat generated by the reduction reaction. The machine chips were mixed with the UF₄/Mg blend prior to loading in the reaction vessel.

Trial runs No. 17 and 18 had chips mixed with the UF₄ and Mg blend in addition to a small amount of lithium metal. The lithium was placed near the bottom of the reaction vessel charge in an attempt to control the point of initiation for the reduction reaction.

From the qualitative evaluation of metal quality and yields, it can be seen that the results were not encouraging. Not only was metal quality and yields poor, the quantity of chips added to the blends were at insignificant levels. As a production technique, it could not keep up with the volume of chips generated from core production. In addition, there was the lingering technical problem of accounting for the titanium added to the derby from the machine chips.

This method was eliminated from further consideration in favor of examining other alternatives with fewer technical problems and with greater potential.

Inductoslag Remelting (ISR)

The ISR method utilizes a salt slag heated by an induction coil. Machine chips can be added to the molten salt where they can melt beneath the salts' protective cover. Specific salt slags may also tend to strip oxide films from chip surfaces and permit molten droplets to coalesce at the bottom of the crucible.

The experimental melt trials that were performed are shown in Table 2. Despite significant time periods of holding the molten chips at temperatures up to 200°C above their melting point, only partial separation between the metal and slag could be achieved. Graphite crucible deterioration was extensive from graphite oxidation at high temperature.

Accurate yield data could not be obtained due to heavy and widely dispersed salt inclusions. Fine machine chips appeared to layer themselves

Table 2. Inductoslag remelting (ISR trials)

<u>Trial no.</u>	<u>Salt used</u>	<u>Weight of salt</u>	<u>Weight of chips added</u>	<u>Heating cycle</u>
1	MgF ₂	13 lb	None	Hold at 1350°C; 1 Hour
2	MgF ₂	16 lb	30 lb	Add Chips to Salt at 1350°C; Hold Temperature, 1/2 Hour.
3	50% MgF ₂ /50% CaF ₂	16 lb	30 lb 5	Add Chips to Salt at 1350-1400°C; Hold at 1400-1450°C 1 Hour
4	50% MgF ₂ /50% CaF ₂	16 lb	30 lb	Add Chips to Salt at 1350-1300°C; Hold at 1400-1450°C, 1 Hour

between small droplets of molten salt. The MgF₂/CaF₂ slag mixture did appear to dissolve and suspend the oxides and the salt did not appear to reach its upper limit of oxide solubility.

Efforts of Worcester Polytechnic Institute (WPI) had preceded the experiments at NMI (Reference Appendix A). Their results looked encouraging and the problems they encountered were attributed to problems of scale. Specifically, it was felt that increasing the crucible size and mass of metal would improve separation of the metal and slag. This did not prove to be the case at least for the 10 in. I.D. x 12 in. high crucible used at NMI.

It was at this point of the contract effort that a program review was held at ARRADCOM. The decision was made to discontinue examining this area of development. Technical feasibility of this recycling method could not be demonstrated within the limits of the current program.

Vacuum Induction Remelting (VIR)

Vacuum Induction Remelting experiments were performed concurrently with the ISR process experiments. VIR was applied to thoroughly dried machine chip briquettes.

The briquettes were prepared in a 250 ton hydraulic press with soft steel tooling that formed a 4 1/2 in. diameter by 1-2 in. thick, compacted briquette. A sufficient number of briquettes were prepared for the melt charges shown in Table 3. Melt numbers UX2037, UX1689 and UX4037 were not cast under this contract. They are included in the table to show additional, earlier attempts using VIR and general progress in technique. The yield of recent attempts (UX5984, UX6426, and UX6523) are fairly good and consistent. Chemical quality typically exhibited a loss of titanium and high iron.

The titanium was most likely floated off during the melting as titanium carbide. This was verified by analyzing the titanium and carbon content in cast billet hot tops. The high iron was later found to have been the result of contamination from soft briquetting tooling. Experiments to evaluate this possibility are discussed later in this text.

Table 3. Chip remelts - briquettes, vacuum induction melted

Chip type (generator)	Weight Melt (lbs)	Date	Charge		Casting as solid recycle derby (%)	Charge as as recycle derby (%)	Crucible & pourcup skull weight (lbs)	C ppm	Ti ppm	Chemistry Fe ppm	O ₂ ppm
			(%)	(%)							
GAU-8	207	UX2037	6/24/80	62.8	(.75 Ti BILLET)	-	79.2	20.8	80/60	.68/.68	-
GAU-8	300	UX1689	5/15/80	100.0	-	-	92.0	-	240/560	.30/.30	-
CNC	1226	UX4037	2/12/81	82.2	GAU-8 SOLIDS	17.8	-	80.0	257	100/110	.64/.65
CNC	949	UX5482	6/16/81	100.0	-	-	67.1	312	300	.50	-
(e) QNC	(d,h) 642	UX5569	6/24/81	100.0	-	(f)	72.1	179	180	.39	- 138
CNC	1200	UX5751	7/28/81	16.7	-	74.4	71.6	-	560	.47	28
QNC	(h,j) 710	UX5984	8/19/81	100.0	-	-	69.0	-	90	.45/.46	42
CNC	747	UX6426	9/30/81	100.0	-	-	72.8	203	90	.54/.55	20
CNC	748	U6523	10/9/81	100.0	-	-	70.0	-	80/60	.54/.54	160/160
										(n)	33/36
										(c)	
										(c)	
										(m)	

NOTES:

- (a) 2-46% Cu
- (b) T.C. #9, LARGE FURNACE
- (c) TIP CHEMISTRY
- (d) FIRE IN CRUCIBLE BEFORE MELT BEGAN
- (e) 12 1/2" ♀ BRIQUETTES IN LIEU OF NORMAL 4 1/2"
- (f) 7.1 LBS Ti ADDED TO MELT CHARGE, ARMY DERBY
- (g) VACUUM DURING MELT: 130-150 MICRONS
- (h) VACUUM DURING MELT: 1500 MICRONS
- (i) BRIQUETTES MUSH, NOT FULLY LIQUID
- (j) EVIDENCE OF OIL IN FURNACE DURING CYCLE
- (k) Ni-28PPM, Al-11PPM, Mg-6PPM, Si-42PPM: TIP SAMPLE
- (l) EXTRUDED ROD CHEMISTRY: Si-83/93, Mg-1PPM, Al-12/14, Mo-4/5, Ni-14/15
- (m) EXTRUDED ROD CHEMISTRY: FRONT/REAR, Si-44/42
- (n) EXTRUDED ROD CHEMISTRY: FRONT/REAR, Si-44/42
- (o) VACUUM DURING MELT: 300-400 MICRONS

Table 3. (cont'd) - Chip remelts - briquettes, vacuum induction melted

Chip type (generator)	U.T.S. (ksi)	Mechanical properties			Hardness (Rc) (8)	Superheat (°C)	Time at temp (mins.)	Hold before pour (mins.)	Pour Temperature (°C)
		Y.S. (ksi)	Elong. (%)	R/A (%)					
GAU-8	-	-	-	-	-	43.2-44.7	1400	5	1340
GAU-8	-	-	-	-	-	1250	5	0	1430
CNC	212-217	127-133	8.0-13.6	12.4-18.3	42.4	1413 (b)	55	5	1403 (b)
CNC	-	-	-	-	-	1438 (b)	20	5	1440 (b)
10 CNC	168-207	111-119	3.1-20.0	1.6-16.8	39.1-41.1	1350 (i)	70	0	1353
CNC	-	-	-	-	-	1247 (b)	30	10	1270
CNC	-	-	-	-	-	1440/1450 (b)	15	5	1330
CNC	-	-	-	-	-	1440/1450 (b)	15	10	1330
CNC	-	-	-	-	-	1440/1450 (b)	15	10	1330 (b)

LEGEND: GAU-8 - 30 mm penetrator chips; CNC - computer numerical controlled lathes

The titanium loss can be adjusted on subsequent remelting by adding additional titanium so that the 0.75 w/o level is reached. The iron, which at the time was considered a serious problem, can only be diluted with low iron virgin stock to bring the iron level down to 80 ppm maximum. Calculations show that virgin stock of 20 - 30 ppm iron content could dilute the iron level in the recycled billets. However, only 8-10% of the dilution melt charge could be composed of recycled material. This rate of recycle addition is not sufficient to keep pace with production scrap volumes. Chemical analysis of non-homogenous derby would make low iron content derby selection difficult. Very low iron derbies are not typical in production and the volume needed for sustaining continuous dilution melt castings are not achievable.

It was these concerns that prevented further consideration of VIR as a viable production recycling process. However, as will be discussed later, the source of the iron contamination was discovered during the Electron Beam remelting efforts. Therefore, in retrospect, reexamination of VIR is now considered warranted.

Electron Beam Remelting (EBR)

Electron beam remelting of chips was proposed at a program review held at ARRADCOM. EBR technology is currently in use as a means of remelt and purification.

NMI examined the state-of-the-art of this technology to determine its suitability for remelting DU machine chips. Discussions were held with personnel from the Department of Energy's Y-12 and Rocky Flats plants to learn of any experiences they may have had with EBR.

EBR is currently used at Rocky Flats to melt and purify unalloyed DU and the DU-6Nb alloy. The technique is quite successful in reducing oxide inclusion levels from solid input material. Researchers at Y-12 have used EBR in past DU and enriched uranium recycling studies. They have found the EBR approach to reduce levels of aluminum, copper, carbon and iron from solid input material.

Y-12 has had limited experience applying EBR to loose and briquetted uranium chips. However, their limited results with certain chips have been very good. Yields greater than 95% have been achieved with excellent quality and good homogeneity. The chips used in this case were coarse turnings that were pickled, rinsed and spun dry prior to melting.

Our initial effort in Electron Beam melting of DU-0.75% titanium chip material was accomplished through the cooperation of the Rocky Flats Plant of the Rockwell International Corporation; theirs being the only facility with the proper equipment and the capability of handling the specific material. In as much as safety considerations prohibit transport of machine turnings, it was necessary that we process the chips by a combination of compaction and extrusion to an acceptable solid form.

The material was thus processed in the following manner. Chips were taken from the finish machining operation of the M774 penetrator and rinsed free of the water soluble cutting oil. Processing then continued through the combination of hot oil and solvent rinses as described in the drying process earlier in the text. The next step in the operation was the cold compaction of the chips in a steel packing sleeve. A load of 250 tons resulted in a briquette 4 1/4 in. in diameter by approximately 1 in. thick. Having once processed a number of these compacts, they were loaded into our standard copper extrusion can. A 1 in. thick copper block was added to the top of the can so that hot compaction of chips could be accomplished by extrusion can upset before the actual extrusion began. An end cap with an evacuation tube was welded to the can which was then pumped down and preheated to assure as much outgassing as possible. The tubing was hot crimped to seal the can (Figure 1).

The billets were heated to 1100°F and extruded to 1.4 in. diameter. The rods prepared in this manner had a density of roughly 94% of the theoretical wrought alloy density. Assuming 100% consolidation of the chips and that the oxides on chip surfaces are U_3O_8 and/or UO_3 (oxide densities of 8.4 and 8.34 gms/cc respectively), calculations indicate the volume of oxide present is approximately 11%.

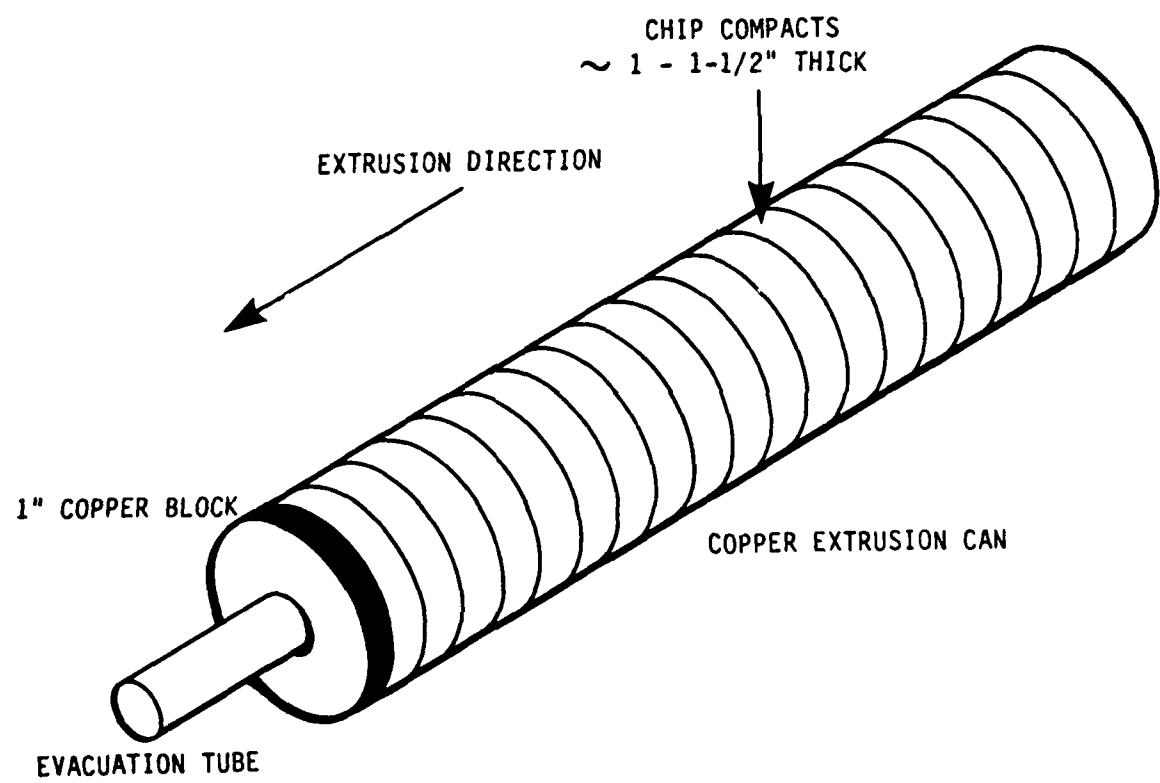


FIGURE 1. EXTRUSION CHIP BRIQUETTE ASSEMBLY

It has been our practice for the purpose of these remelt experiments to remove the copper cladding on the extruded rod by machining .

Rods prepared in this manner were shipped to Rockwell International for remelt.

The furnace at Rocky Flats is a Leybold-Heraeus system having three 60 Kw guns as a power unit. This particular furnace is equipped with a side chamber into which the rods were charged and fed. A total of 68 lbs was thus processed resulting in a final cast billet 4 in. in diameter by 11 in. long. The 11 in. length includes 2 in. contributed from an unalloyed DU starter. This block was placed in the water cooled continuous casting mold assembly prior to melting the extruded machine chips.

The melt was sectioned and subjected to both chemical and metallographic examination, the results of which may be found in Tables 4 and 5 and Figures 2, 3, 4, 5, and 6.

Since this was an experimental melt where the optimum EB melt cycle was unknown, some adjustments in power settings and beam position were made by the operator throughout the procedure. Consequently, these changes appear to have manifested themselves in some differences in chemical uniformity as shown in the analysis. It is generally considered that these differences have no significance at this point in time.

With the exception of iron and titanium levels, the chemical quality of the ingot was well within the specification presently required for core production.

It should be pointed out that the steel packing die used in the preparation of the briquettes (for this effort and for the VIR experiments) was suspected as the source of the iron contamination. The chips were quite abrasive resulting in much galling of the briquetting tooling.

Table 4. E. B. billet chemistry

Sample no.		Ti (%)	C (%)	Fe (ppm)	Ni (ppm)	Si (ppm)	Cu (ppm)
Top	1-A	0.52	0.007	129	9	30	14
	1-B	0.60	0.004	129	11	44	15
	1-C	2.19	0.005	270	9	42	6
	2-A	0.49	0.006	134	12	33	27
	2-B	0.49	0.005	124	13	37	23
	2-C	0.54	0.005	142	12	38	25
	3-A	0.53	0.006	106	20	30	11
	3-B	0.53	0.005	109	20	49	10
	3-C	0.54	0.003	112	18	43	11
	4-A	0.55	0.006	112	10	90	20
Bottom	4-B	0.52	0.005	112	8	104	36
	4-C	0.48	0.005	89	8	92	17
	Surface #2	0.50	0.005	160	23	52	11
Surface #4		0.33	0.007	136	15	123	8

Table 5. E. B. billet

Spectrographic analysis ppm

Element	Spec. No.			
	1	2	3	4
Ba	1	1	1	2
Cr	1	1	1	1
Co	1	1	1	1
Va	4	4	4	5
Mg	1	1	1	1
Al	71	2	3	5
Cu	8	21	12	14
Ni	11	14	20	11
Mn	2	4	6	8
Fe	92	133	106	120
Si	45	45	45	95
Zn	1	1	1	1

H

Oxygen

1	7400	Porous	1-X	0.7
2	13		1-Y	7.0
3	14		2-X	0.6
4	13		2-Y	0.5
			3-X	0.9
			3-Y	1.0
			4-X	0.8
			4-Y	0.8

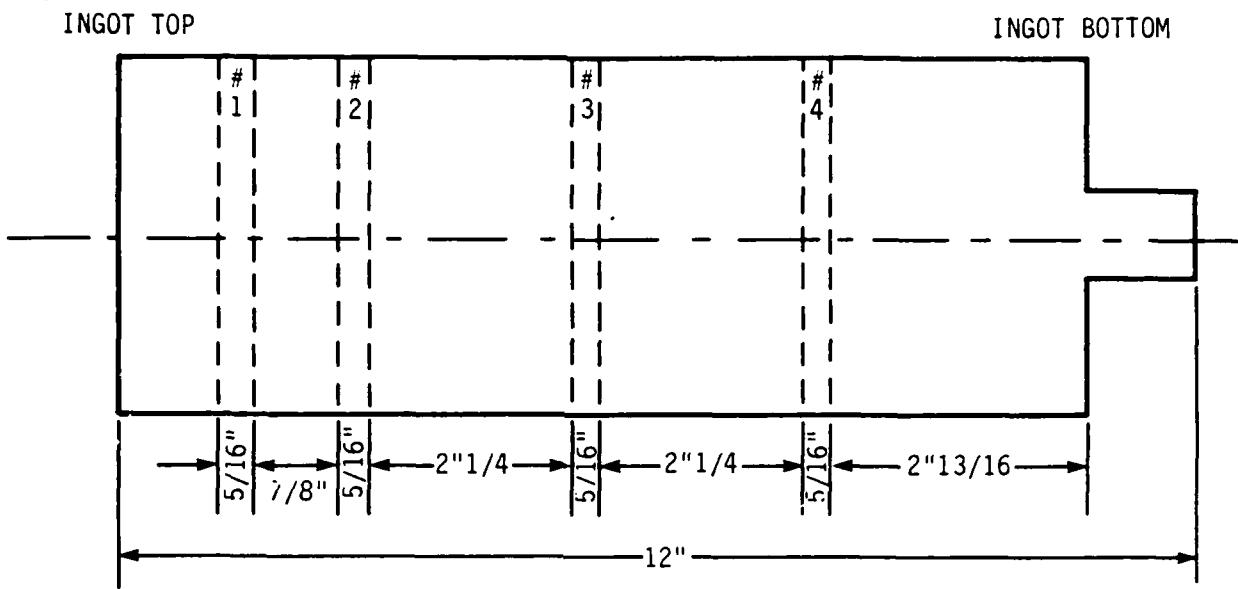


Figure 2. Sections of Electron Beam Billet

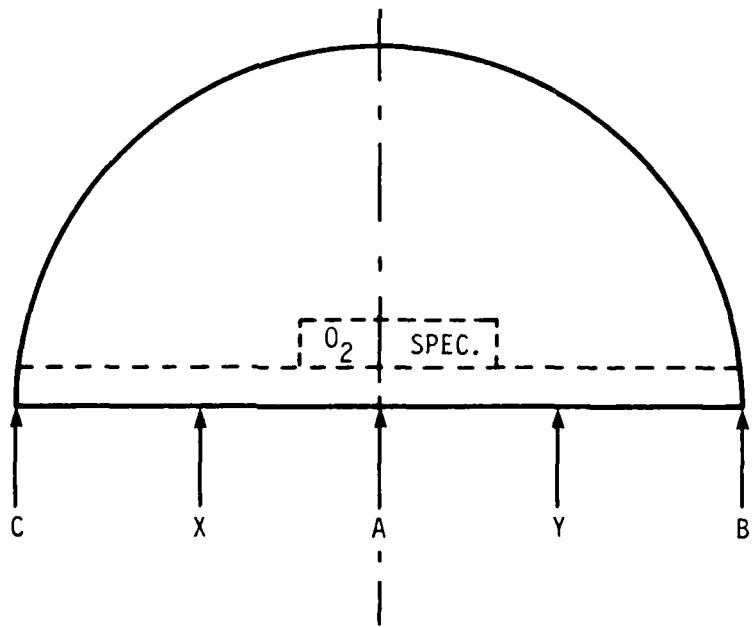


Figure 3. Specimen Locations

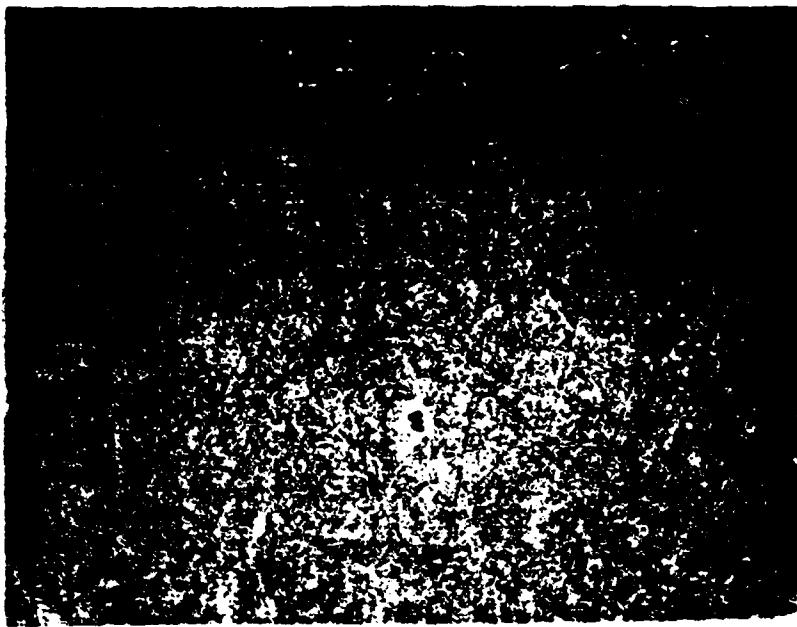


Figure 4. As polished 1st E.B. melt, 100X

The as polished photomicrograph shows generally good clean material. The few very small spots are carbides.

In Figures 5 and 6, the etched condition shows large equiaxed grains, well defined with a basic acicular structure.

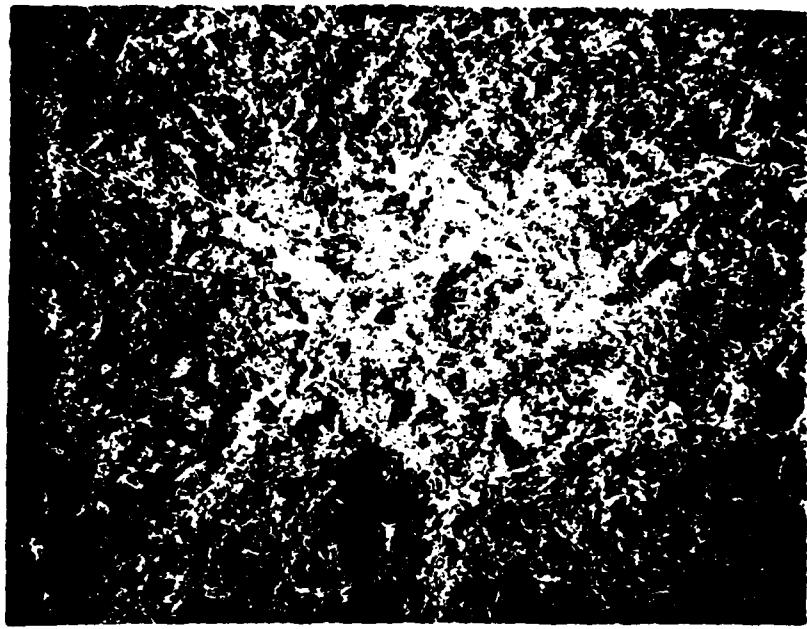


Figure 5. Etched 1st E.B. melt, sec. 2, 100X

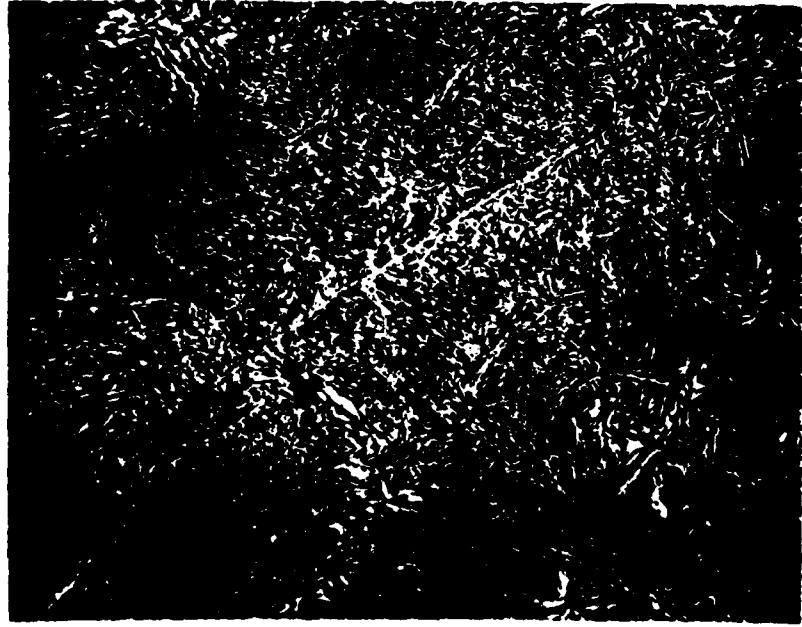


Figure 6. Etched 1st E.B. melt, sec. 4, 100X

Following discussions with ARRADCOM personnel, it was decided that a follow-up experiment specifically designed to address the problem of iron contamination was appropriate. We therefore designed and fabricated a sleeve and end plates for a briquetting die entirely from DU-0.75 Ti alloy, thus minimizing any chip contact from a possible iron contaminating source. Another source of iron contamination was identified as a result of chip conveyor wear at the CNC lathes. Chips were passed over a large magnet to remove the small steel filings prior to briquetting for the second remelt attempt.

New chip material was prepared, compacted into briquettes and extruded in the same manner as the original experiment. Chemical analysis, specifically for iron in the individual rods, prior to remelt are reported in Table 6.

Arrangements were made to remelt this material using the electron beam furnace at Rocky Flats.

Some difficulties were experienced with the vacuum system on the Electron-Beam furnace with the result that it was not possible to pump down to the range of 10^{-4} Torr or better as had been achieved on the first melt. At this point, time was a factor and since our prime concern was only to address the iron problem it was decided to proceed with a much lower vacuum level in the chamber. The rods were melted to billet form and subsequently returned for examination at NMI.

As a matter of comparison, we analyzed for titanium, carbon, hydrogen and oxygen as well as the iron. Two slices were cut from the billet in the same relative positions as numbers 2 and 4 as shown in Figure 2 of the first melt. The results are listed in Table 7.

In all cases of sampling for iron contamination in the extruded chip rods, the detected iron levels are suspect. A small iron chip would not contribute to detected iron levels unless it was present in the tested sample. It is therefore impossible to conclusively prove that the EB remelt approach has resulted in a reduction of the iron content in the final reclaimed ingot. Recent experiments at Rocky Flats Plant using the EB approach to fabricate a DU-7Nb alloy has demonstrated a refinement capability. VIM homogenize DU-7Nb alloy with a carbon content of 150 ppm was EB remelted and resulted in a carbon level of 50 ppm. Those individuals with experience remelting DU within the DOE make similar claims for iron refinement. Hard data has been difficult to obtain.

It is felt that the poorer vacuum observed in the second EB remelt experiment was responsible for the higher values of both the hydrogen and oxygen. This condition, along with an EB power problem experienced during the second melt, is manifested in the degree of porosity observed in the second billet that was not present in the first EB melted billet. However, it has been demonstrated that material prepared by this method is of a quality suitable for recycle as defined by the present core specifications. The titanium level can be raised to the proper value upon a subsequent VIM remelt.

Table 6. Iron analysis of briquetted and extruded rods
for 2nd electron-beam experiment

<u>Rod no.</u>	<u>Iron (ppm)</u>
1	19
2	22
3	24
4	18
5	28
6	26
7	24
8	29
9	18

Table 7. Analysis of 2nd electron-beam billet

Section 2.	Ti	0.52%
	C	0.005%
	Fe	47 ppm
	H	3.3 ppm
	Oxygen	53 ppm
Section 4.	Ti	0.47%
	C	0.002%
	Fe	38 ppm
	H	3.5 ppm
	Oxygen	17 ppm

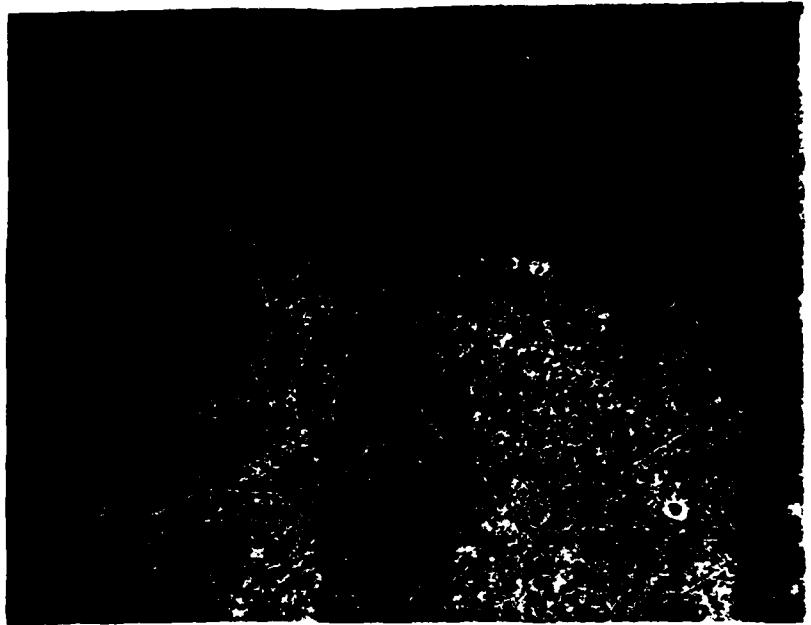


Figure 7. As polished 2nd E.B. melt, 100X

There is no discernable difference in the as polished condition between the first and second melts. We again see a few minor carbides.

In the etched condition, as shown in Figures 8 and 9 taken from billet positions relative to sections 2 and 4 as shown in Figure 2, we have a structure identical to the first melt.

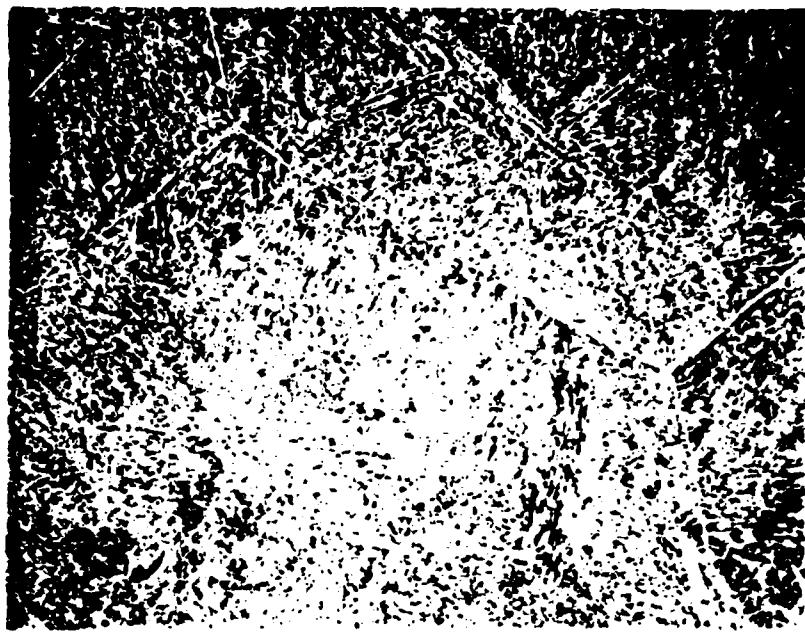


Figure 8. Etched 2nd E.B. melt, sec. 2, 100X

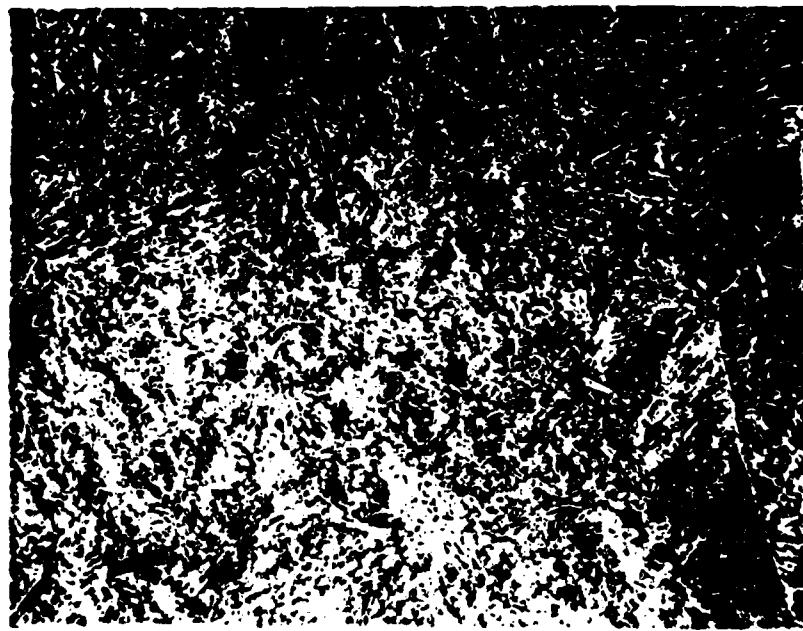


Figure 9. Etched 2nd E.B. melt, sec. 4, 100X

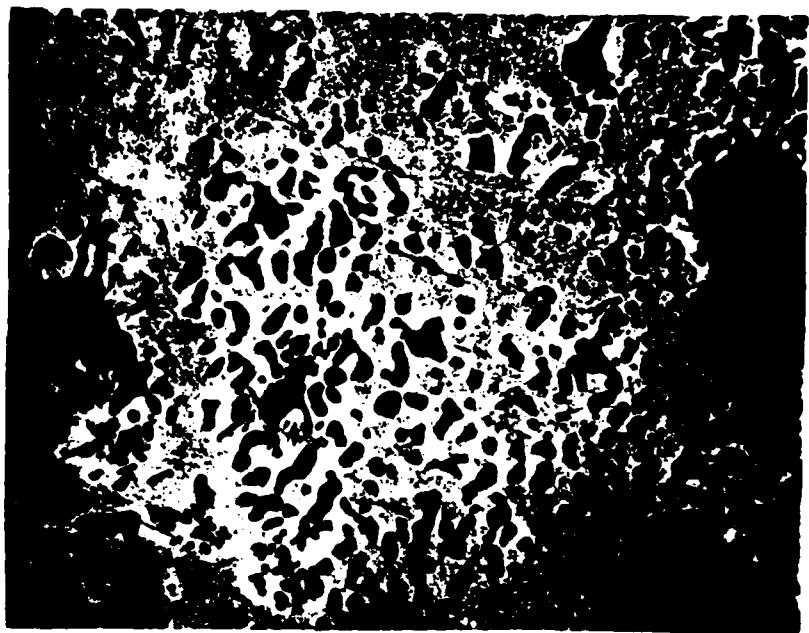


Figure 10. 100X

For purposes of illustration, Figure 10 shows typical hot top slag as was observed in both electron beam melts. What is observed is a combination of oxides and carbides. The large dark areas are voids.

DISCUSSIONS AND CONCLUSIONS

Having completed the recycling approaches as described in the program, we should now like to address each of the procedures in their respective order.

Conversion of Machine Chips to UF₄

The complications involved as a result of the generation of uranyl fluoride (UO₂F₂) in the reaction to UF₄ did not justify continued effort in this approach. While some degree of success was achieved, the decision was made that the time frame in which the program was to be conducted and the funding limitations of the overall study did not warrant an extreme effort at the expense of other procedures which offered greater potential. Therefore, further consideration of this approach was dropped entirely.

Co-Reduction of Chips in the UF₄/Mg Reaction Vessels

Early on in this portion of the study, the laboratory size reduction attempts indicated rather clearly that the potential levels of chip additions would be so low that one could not consider it a viable technique. The technical problems of titanium additions to derby from the machine chips only served to add another complication. It was therefore felt that other approaches being considered should take precedence.

Inductoslag Remelting

A most intensive effort was mounted in the inductoslag study. A masters thesis was funded at Worcester Polytechnic Institute (Appendix A) to explore various approaches to salt combinations which would strip the oxide coating from the chips and permit the recovery of clean metal.

On a strictly laboratory scale the results suggested potential for this approach. Of those problems that did arise, it was felt that if the trials were increased in scale the greater material mass would lead to improved metal/slag separation. Regretfully, this did not appear to resolve the issue. The effect of the layering of the chips in the molten salt could not be overcome irrespective of long holding times at highly elevated temperatures. Since only partial separation was achieved, we had to conclude that the demonstrating feasibility of this approach was outside the scope of the program.

Vacuum Induction Remelting

Examination of vacuum induction remelting was conducted simultaneously with the inductoslag effort. Our approach in this case required the briquetting of chips in order to achieve adequate furnace loading from which we could properly draw significant yield information. As is contained in the body of this report, yields in the range of 70% coupled with obvious contamination of carbon and iron did not at that time give strong promise of an ultimate solution. As was noted, there was a decrease in titanium on recovery. This fact is really of no consequence when we know that with an additional remelt, this loss can easily be brought back into specification. Other possible contaminants pose the primary concern. Remelting by VIM processing in no way allows us to "clean" the metal and if anything, further exposes the metal to additional carbon contamination.

Preventing iron contamination at the briquetting step can be readily accomplished by utilizing DU tooling. Removing contamination from the chips by magnetic separation is not a simple or absolute solution to iron contamination prevention. Once DU is contaminated with iron and vacuum induction remelted, it cannot be removed by VIR, only diluted.

While the iron problem has the potential to be overcome, carbon contamination, on the other hand, does not. Carbon will progressively accumulate in the material as pickup from the crucible wall as the chips are remelted for recycle. In all likelihood, at some point in time the carbon will reach an unacceptable level where some material will have to be scrapped outright to purge the system of high carbon material. This situation has been experienced at the DOE's Y-12 facility where a DU-6Nb recycle program has been in place for several years. Solid scrap has contributed to a continuing rise of carbon contamination. It is not known what proportions of scrap are recycled with virgin stock or how many generations of material were reprocessed before the situation became apparent. It does demonstrate the inevitability inherent in such a process.

Not only must we concern ourselves with the quality and quantity of the recovered material; equally important is the cost effectiveness of the total recovery process. It is, after all, the purpose of the recycle program to reduce the cost of penetrator production.

In this study, we had to briquette out of necessity a labor and cost intensive approach. Graphite tooling is also required for this operation, material which has a limited useful life and one which contributes to carbon contamination. There is, of course, the added waste disposal costs associated with pyrophoric casting skulls and graphite tooling. All of these factors will therefore play a most important role in weighing one procedure against another.

While the above considerations detract from the cost effectiveness of VIR, it is also true that there was a 70% yield from the process. Also, we were not able to examine every facet of the process within the confines of this program. Therefore, it is believed that a more sophisticated and detailed undertaking in VIR is justified.

Electron Beam Remelting

Electron beam technology is presently being used in the metal industry where there is not only concern for the recovery of various special metals but where the quality of such materials is of the utmost importance. It was this very fact which prompted our proposal to ARRADCOM suggesting we examine the technology in detail to determine the benefits from such an approach in this program.

There are production sized electron-beam furnaces presently using a water cooled copper hearth and mold. The copper tooling is reusable many times before its useful life limit is reached and it has the further advantage of not being a source for reclaimed metal contamination.

The typical furnace design allows for the continuous casting by ingot withdrawal from a water cooled mold. There are established techniques by which it is possible to remove oxides before entering the casting mold, thus assuring a clean melt. Most important is the capability of this system to clean the processed metal. This is essentially done by controlling the melting cycle in such a manner as to combine the heat of the electron beam itself with the vacuum to effectively distill off certain contaminants.

On the negative side, it has been suggested that distillation of high quantities of contaminants would degrade vacuum levels and result in contamination of diffusion pumps. However, limited experiments at Rocky Flats Plant have had no effect on the existing vacuum pumps, and it is believed engineered capture mechanisms can be employed to prevent this potential problem. Vacuum systems for "dirty" environments are designed for pumping capacities which can meet forecasted outgassing levels. Unfortunately, the validity of this concern relative to distillation of uranium oxide contaminants can only be determined in the production mode after a \$3 million capital investment.

In our particular experiments at Rocky Flats we were only able to melt the fabricated rods to billet form by drip melting; a hearth system was not available. In discussions with the staff at Rockwell International, it was indicated that should we have the opportunity to properly establish a more desirable EB melting process, namely a longer holding time in the liquid stage, we could produce an even more chemically clean material. We suggest that it is this factor above all others considered which would call for an in depth study to establish the EB process parameters which, it is believed, would result in class A recycle material.

SUMMARY

Of all the processes investigated, only VIR and EB are considered worthy of pursuit. EB is considered more likely to succeed as it provides a product in which impurities are refined. It also has less recurring costs as there is no disposable tooling (graphite crucibles) to be disposed of. Unfortunately, capital investment is extremely high and, consequently, this represents a high risk approach.

VIR, on the other hand, has a lower relative yield (70 vs 95%) and has higher recurring costs. It is readily integratable into the current manufacturing process and has a relatively low capital investment.

APPENDIX A

THE RECOVERY OF MACHINED URANIUM CHIPS

by
Hsi-Chieh Tsai

A Thesis

Submitted to the Faculty
of the

WORCESTER POLYTECHNIC INSTITUTE

in partial fulfillment of the requirements for the
DEGREE OF MASTER OF SCIENCE
IN
MATERIALS ENGINEERING

ABSTRACT

A method was found to recover more than 80% of the uranium from severely oxidized depleted uranium machining chips. Under inert atmosphere, the chips, coated with salts such as (MgF_2 , $CaCl_2$, $BaCl_2$) were stripped of their oxides by a fluxing slag which separated from the metal by flotation.

Optical microscopy, x-ray diffraction, scanning electron microscopy, energy dispersive x-ray analysis, and chemical analysis indicated that the resultant metal was of purity comparable to that of the original metal from which the chips were produced. The carbon content increased slightly because of the graphite crucible. The results are considered in terms of potential scale-up for industrial use.

ACKNOWLEDGEMENTS

I wish to take this opportunity to express my sincere appreciation to Mr. Mark Walz of Nuclear Metals, Inc. for his support in conducting this research, and to thank Profesors Roger J. Austin, Alfred A. Scala and Ronald R. Beiderman for their invaluable advice throughout the program. Finally, I would like to thank Mr. George Schmidt and Giacomo Ferraro for their help toward the completion of this project.

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INTRODUCTION

Nuclear Metals, Inc., (NMI) of Concord, Massachusetts, manufactures penetrators of Uranium-Titanium alloy containing 0.75% Ti. The uranium used is mainly U-238, also known as "depleted uranium" because of its low-level radioactivity. In the manufacturing processes, machining chips result which may be discarded only in specific ways prescribed by the Environmental Protection Agency and the Nuclear Regulatory Commission. The currently approved disposal procedure involves encapsulating the chips in concrete within steel drums and shipping to designated burial grounds. The expense of this disposal process has motivated this research.

Conceivably, the "waste" depleted uranium could be reused. If so, not only would there be a materials saving, but the costly disposal process would be eliminated, or at least substantially reduced. How does one save the metal from becoming oxide so that it may be returned to useful purposes as inexpensively as possible? With this in mind, the objective of this research was to identify a promising recycling process. It would be selected based on chemical and metallurgical analysis of the depleted uranium chips and the oxide layer which so readily formed.

The primary means of determining process feasibility was the comparison of chemical composition and metallurgical structure of the depleted uranium before and after the process.

Hazards of U-238

- 1) The material is slightly radioactive. Exposure should therefore be monitored via appropriate film dosimeter.
- 2) The toxicity of U-238 derives from its high atomic number (and thus density). The body has difficulty eliminating it (e.g. lead); thus

the importance of not taking the material internally.

3) Because of its propensity to oxidize, U-238 is dangerously pyrophoric. Actually it is this aspect of the material which not only is a hazard but also is directly related to the object of this research.

Appendix G provides a more detailed discussion of the hazards associated with U-238.

Characterization of the Alloy

The alloy under investigation has the following composition (15):

Titanium	0.75 wt%
Carbon	80 ppm. max.
Iron	80 ppm. max.
Silicon	60 ppm. max.
Magnesium	30 ppm. max.
Uranium-238	remainder

The unique nuclear properties of uranium coupled with its high atomic number (high density) suggest several applications. As an engineering material, however, uranium has several undesirable properties (A-1). It has a relatively low yield strength and rapidly loses its strength with increasing temperature. Also, it oxides rapidly and requires special treatment (such as plating) for protection. Alloying is the most effective way of enhancing the engineering properties of uranium. Of the many uranium alloys, those with less than 1 weight percent titanium appear most promising for the development of a high strength, corrosion resistant alloy with minimal degradation of the properties. The superior mechanical properties result from the fact that the alloying addition is highly

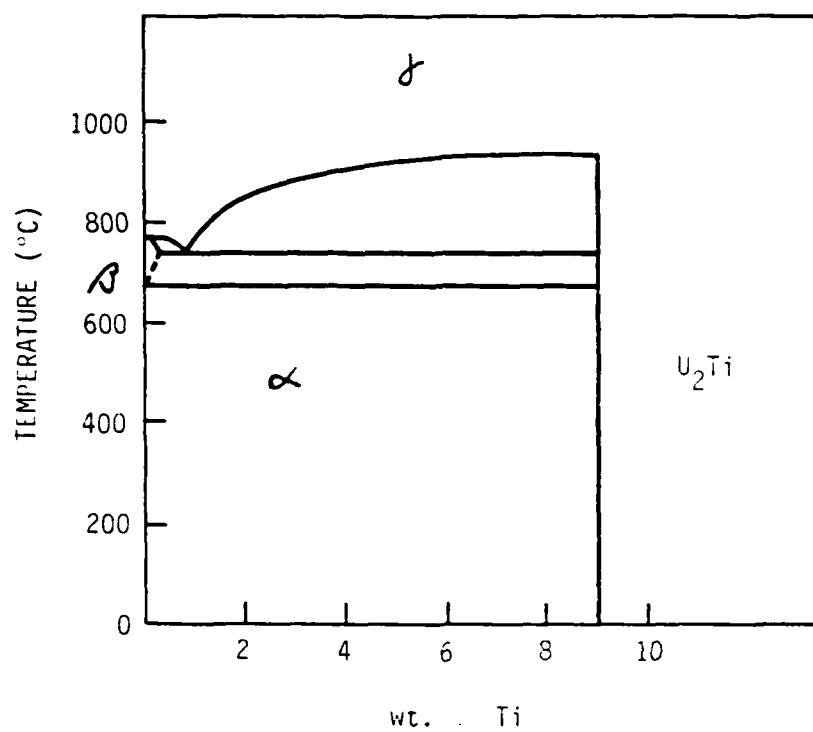


Figure A-1. The uranium-titanium phase diagram

soluble in the high temperature gamma(γ) phase (see the phase diagram shown below) and almost completely insoluble in the low temperature alpha(α) phase. Upon quenching from the γ phase, the addition of alloying elements will suppress the formation of the α phase and produce metastable α' phase alloys. Aging of this alloy at some temperature, therefore, often produces higher strength due to the thermal decomposition of the metastable phase which is normally called precipitation hardening phenomenon.

The Heat Treatment of U-Ti (0.75%) Alloy

On slow cooling, the α phase decomposes via a diffusional phase transformation, the room temperature product being a two phase structure of α phase (almost pure uranium) plus either an intermediate or an alloy-rich phase (see Fig. A-2). The strength of this slowly cooled alloy is only modestly superior to that of unalloyed uranium.

On rapidly cooling from the α phase field, the diffusional transformations are suppressed and the α phase transforms to a variant of the α phase via a martensitic transformation (the product is commonly termed α', a distorted orthorhombic structure). Since this transformation is diffusionless, all of the alloying element is retained in solid solution in the product phase. The α' phase (see Fig. A-3) is therefore supersaturated with alloying elements and is thermodynamically unstable.

Despite the instability, these phases do not decompose at room temperature because of the prohibitively low rate of diffusion. If the temperature is increased however, diffusion becomes more rapid and the α' structure tends to transform to the equilibrium phases. The effect of aging on the yield strength of U-0.75% Ti is shown in Figure 4.

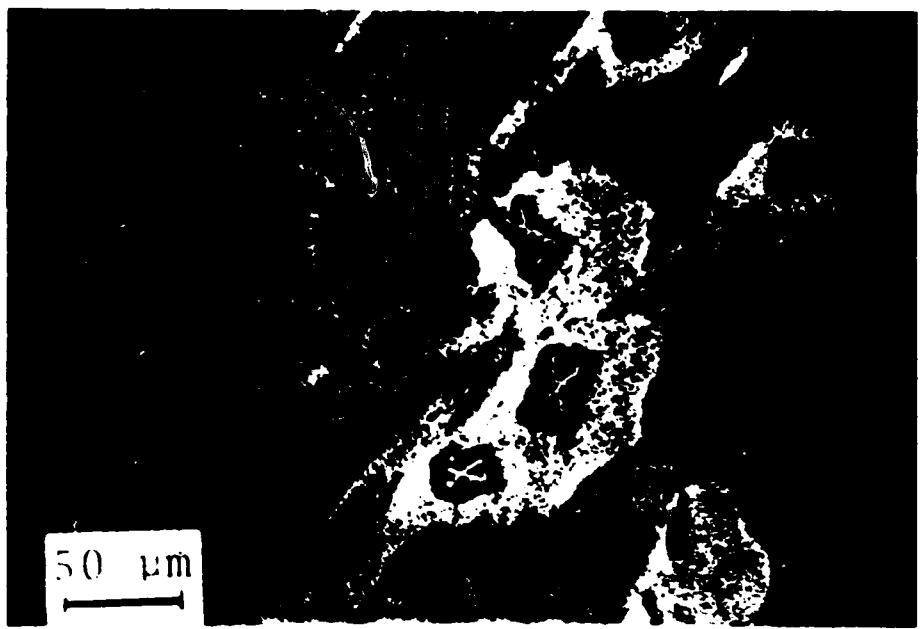


Figure A-2. α -U and intermediate alloy rich phase
(bright field, 200X)

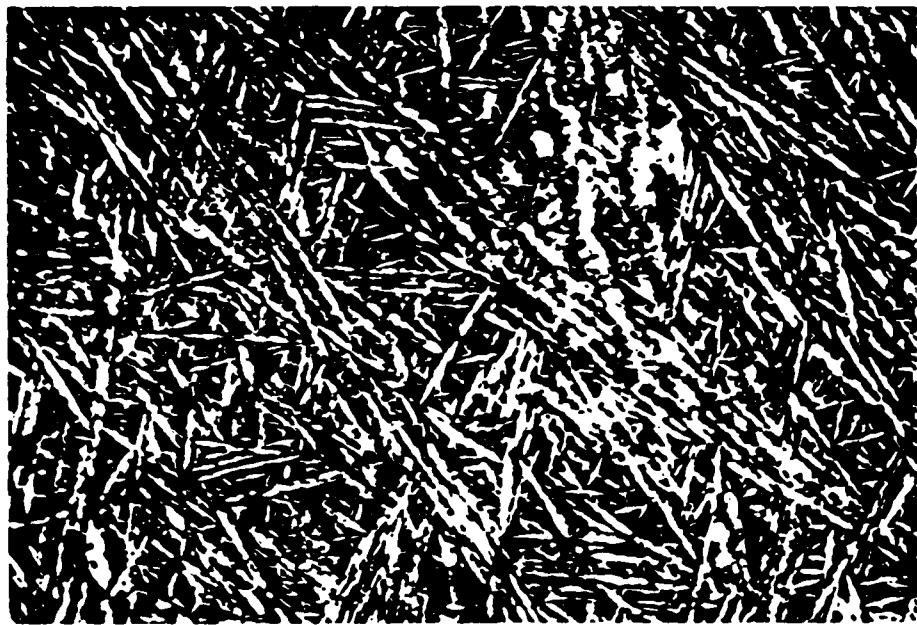


Figure A-3. Acicular martensitic structure in as-quenched
U-3/4% Ti alloy

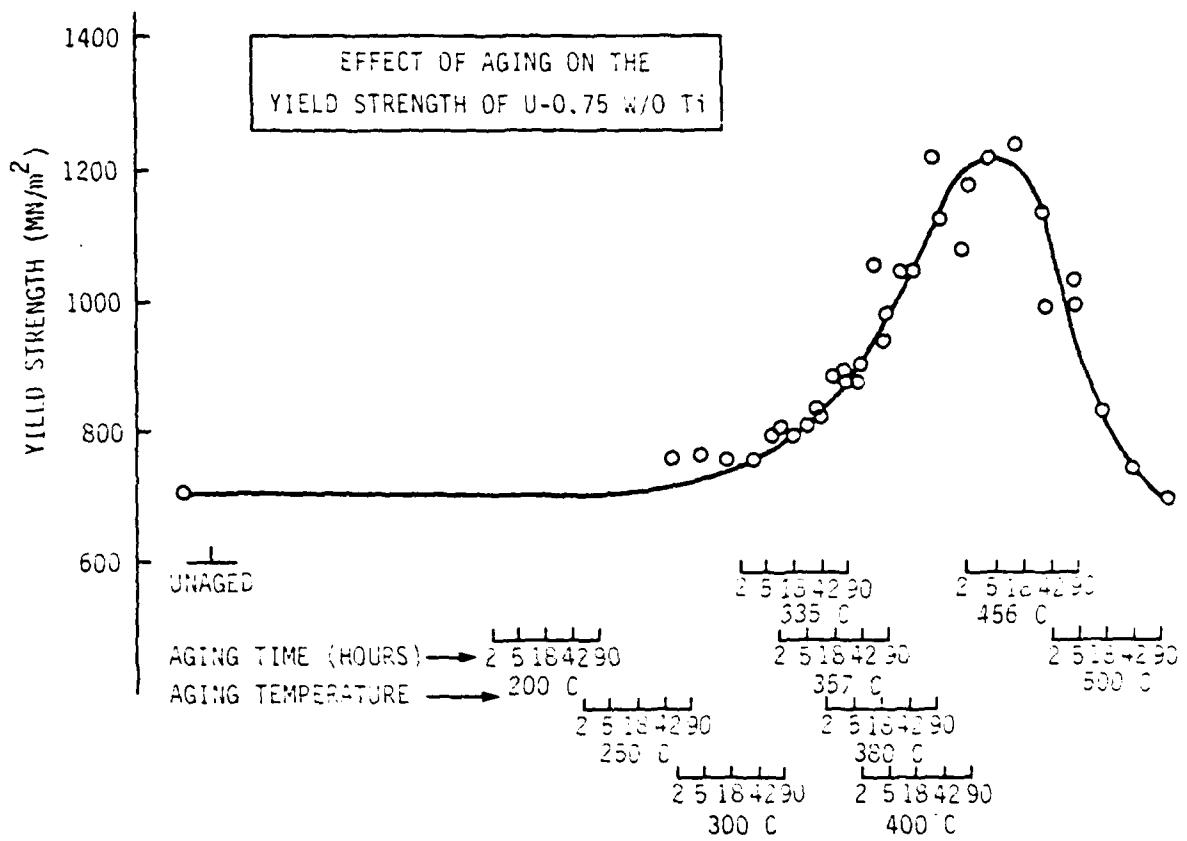


Figure A-4. Aging behavior of U-3/4% Ti

EXPERIMENTAL PROCEDURE

The Melting Processes

The general practice of melting depleted uranium chips has been generally unsatisfactory because of the propensity of the metal to oxidize.

The adherent, rigid layer of uranium oxide, once formed on the surface of the chips, tends to act as a "skull" surrounding the molten metal. Heated even to temperatures several hundreds of degrees above the melting point of uranium, the metal has difficulty coalescing and forming a usable mass.

Several trial methods, using various salts (fluxing agents), were used in the early stages of this research. These methods will now be described briefly to reveal typical features of the results progressing from the early stages to the final selections.

Open-Air Melting

1. The clean, dry uranium chips were placed in crucibles of graphite (the induction furnace susceptor) and were covered by powdered magnesium fluoride. Above 400°C, fumes due to the burning of the chips were emitted from the crucible. It was concluded that atmospheric oxygen, which existed in the chips and the interstices of the chip mixture, severely oxidized the uranium.
2. Clean, dry uranium chips were dropped into molten magnesium fluoride exposed to air. Copious fumes together with fire developed even before the chips were fully immersed in the molten salt. The pyrophoricity and fineness of the chips demanded a protective atmosphere of some kind.

3. Clean, dry and compressed chip pellets were coated with powdered magnesium fluoride and were quickly pushed by a ceramic rod into the molten salt (MgF_2). After holding at $1350^{\circ}C$ about five minutes, sparks started to occur over the surface of the melt indicating that oxidation of uranium chips was proceeding. Spreading or pouring more salt over the melt only temporarily prohibited the sparking phenomenon. This method, however, did yield a metallic product. About 30% of the initial chips were recovered.

4. The same chips as used in method 3. were placed in the crucible and covered with salt up to $7/8$ the depth of the crucible. A cover was placed over the crucible and a magnesium oxide pipe (1/8 inches diameter) transmitted argon to the crucible interior. The temperature was held for 25 to 30 minutes at $1350^{\circ}C$. After breaking the salt away, clean bright metal bulk appeared in the bottom along with some slags mixed with metal at the top of the melt. The recovery yield was still about 30%.

5. Argon was flushed over the premelted salt. Then pelletized chips (see Appendix B, Figure B-3) were dropped and quickly pushed below the surface of the salt bath. Secondly, a cover was placed on the crucible to isolate it from the atmosphere. The recovery yield rose to 65%. Attention was then focused on improved atmospheric control.

Vacuum Induction Melting

After pumping the box (refer to figure D-2) to less than 15 inches mercury, it was backfilled with argon (or helium). Three such purges were done to minimize oxygen contamination. During the melting process, a slight positive pressure of argon was maintained and the box was continuously flushed with new argon.

The melting operation could be seen clearly through a sight glass and the temperature was determined as before, by the thermocouple inserted into

the crucible (see figure D-2). The top hole (1/8 inches) was also used as the exit hole for the argon, thereby preventing overpressure.

The charging assembly was composed of glass pipe (44 mm) sealed by a piston manually operated from outside the furnace. The pellets, placed in the glass pipe before closing the furnace for gas purging and heat-up, were pushed into the crucible after the salt had completely melted (see figure A-5).

The recovery yields from the use of this vacuum chamber ranged from 80 to 90%.

ANALYSES

Metallography of Uranium

Uranium metal exhibits three crystalline forms. The alpha phase which exists up to a temperature of 660°C has an orthorhombic crystal structure. The beta phase, which exists between 660 and 760°C, has a tetragonal crystal structure. The gamma phase, which exists between 760°C and the liquid, has a body centered cubic structure (5). The alpha uranium with which we are concerned, (because of its orthorhombic structure), has a high degree of anisotropy and thus is very sensitive to mechanical deformation. This makes the preparation of a metallographic surface by mechanical techniques extremely difficult. In addition, uranium is very chemically active and it has, therefore, been difficult to devise an etchant which will delineate grain boundaries satisfactorily. The anisotropy of the orthorhombic alpha structure provides for distinctive metallographic studies via polarized light (5); however, rapid surface oxidation diminishes the likelihood of a clearly resolved microstructure after air exposure of several hours. In addition, polarized

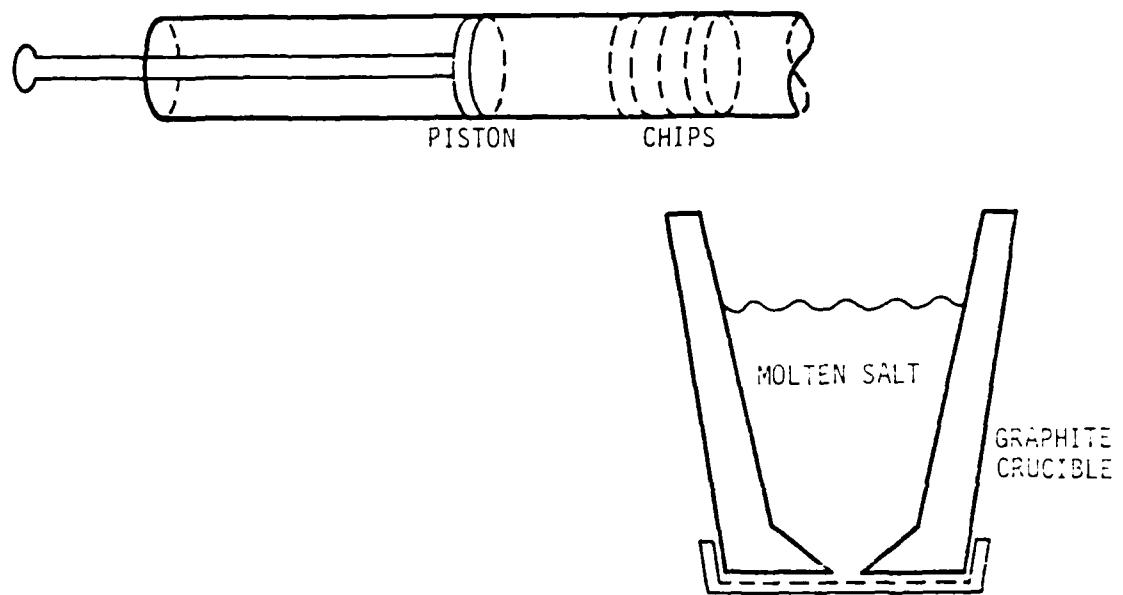


Figure A-5. The charging assembly inside the vacuum induction furnace

light is not generally applicable for the study of inclusions; a major concern in this project.

Chemical and Energy Dispersive X-ray (EDX) analyses indicate that the recycled metal is better than 97% pure and X-ray diffraction shows the U-238 to have the same structure as virgin material (U-0.75% Ti).

Metallographic identification of the inclusions in uranium, however, was a more difficult task; in fact, it was the major work during the latter half of this research.

Uranium carbide which formed during melting from the carbon contamination of the graphite crucible, magnesium fluoride or other slag type entrapped in the melt, and uranium dioxide, were the most common inclusions found in our samples.

Uranium carbide, observed primarily near the melt surface, appeared as metallic white (optically) and typically rectangular or cubic (figure A-6). The carbides which were analyzed by the EDX, however, did show the existence of alloying ingredient. As shown in Figure A-7, those are titanium, silicon, iron, aluminum, copper and zinc. Further, the carbides, when etched with a 50% HNO₃ water solution, did not show any color change (5). Thus it seems that the alloying elements react with the crucible or react with the carbon present in the virgin charge, tie up as carbides, and float to the top of the melt.

Dendritic titanium-rich inclusions were typical, especially in the upper portion of the solidified alloy. In the lower portion, the titanium dentrites were rarely observed owing to the considerable difference in density between Ti and U (Figure A-8).

Slag inclusions were the most abundant and could be identified merely by appearance. They are large, round, and dark particles in the center of the specimen and appeared as continuous agglomerates near the top of the

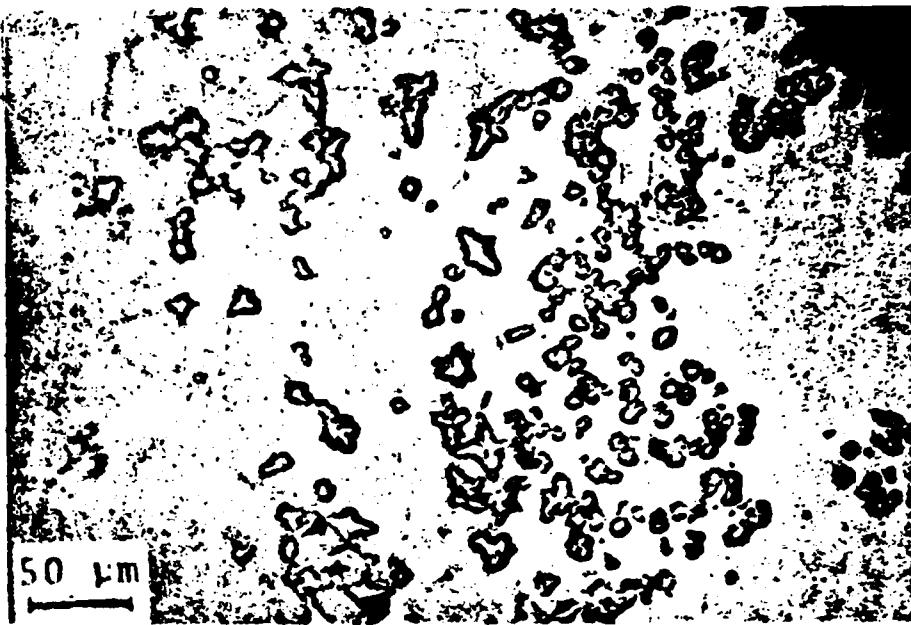


Figure A-6. Uranium carbides observed near the melt surface (metallic white) (bright field, 200X)



Figure A-7. EDX analysis of the carbides shown in Figure 6



Figure A-8. Dendritic titanium rich inclusions (by SEM, 500X)

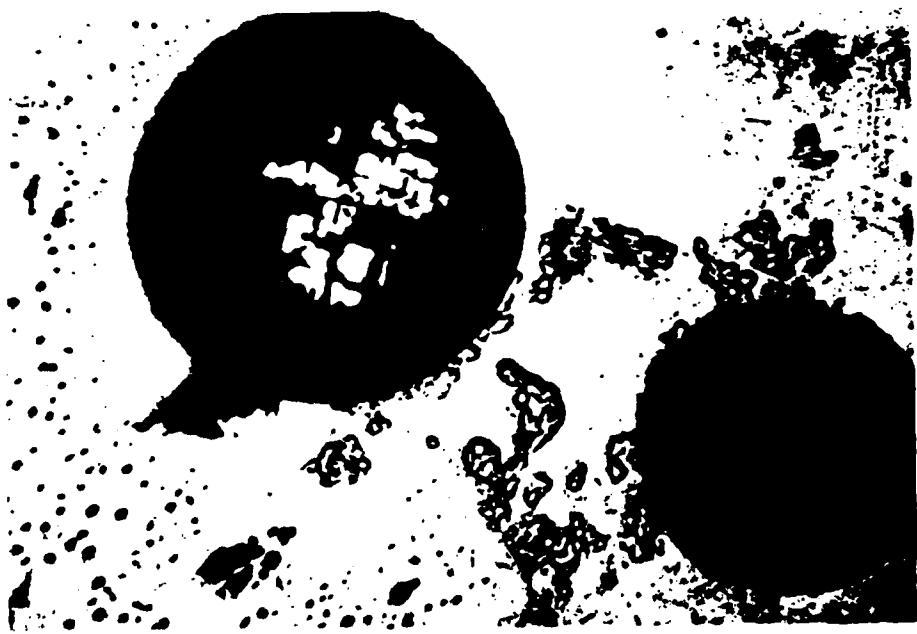


Figure A-9. Slag inclusions found in the center of the melt
(bright field, 200X)

melt. A typical example of the magnesium fluoride inclusions is illustrated in Figures A-9 and A-10. In many cases, identification may be made immediately after mechanical polishing.

Uranium dioxide inclusions also were copiously found in this study because of the severe oxidation in the chips. When present, they were relatively easy to identify by appearance. These inclusions are dark in color and exhibit a generally rounded shape but much smaller compared with slag inclusions. (Figure A-11).

Energy-Dispersive X-ray Analyses

Several as-cast metals, after polishing, were examined under a Scanning Electron Microscope and subjected to Energy-Dispersive X-ray chemical analysis.

Matrix, grain boundary, and each type of inclusion were analyzed by the bulk scan or by spot position technique as follows:

1. Uranium matrices show 100% uranium.
2. Inclusions of round shape show that they are composed of uranium and magnesium. Since magnesium comes from the salt (magnesium fluoride) and fluorine and oxygen can not be detected by EDX, we can suppose that those round dark particles consist of magnesium fluoride and uranium oxide which are soluble during the melting (Figure A-12).
3. Small angular inclusions are composed of 0.64% Si, 8.77% Fe, and 90.59% U. (Figure A-13).
4. Small angular inclusions which differ from 3. are composed of 9.07% Al, 7.63% Fe and 83.31% U. (Figure A-14). Combining with 3 and 4, it seems possible that the alloying elements, which already existed in the metal

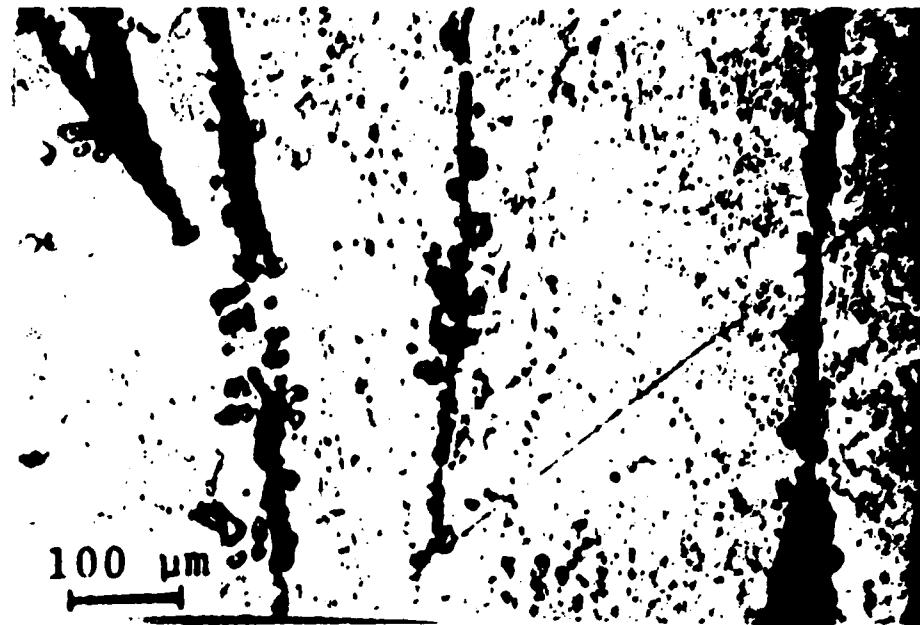


Figure A-10. Continuous slag agglomerated near the top of the melt
(bright field, 100X)

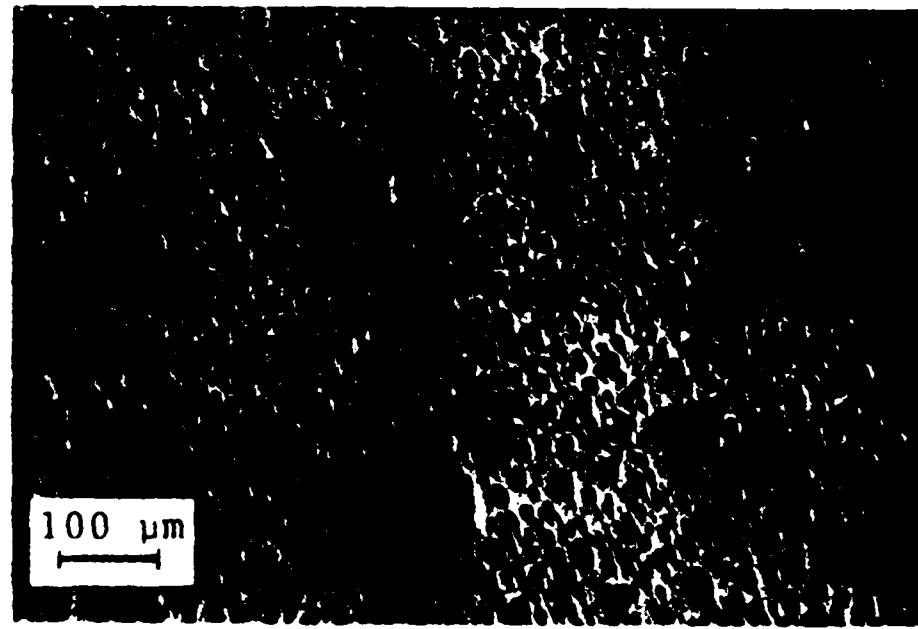


Figure A-11. Uranium dioxide inclusions (as-polished;
bright field, 100X)



Figure A-12. Magnesium fluoride (round shape) identified by EDX
(as-polished, 200X)

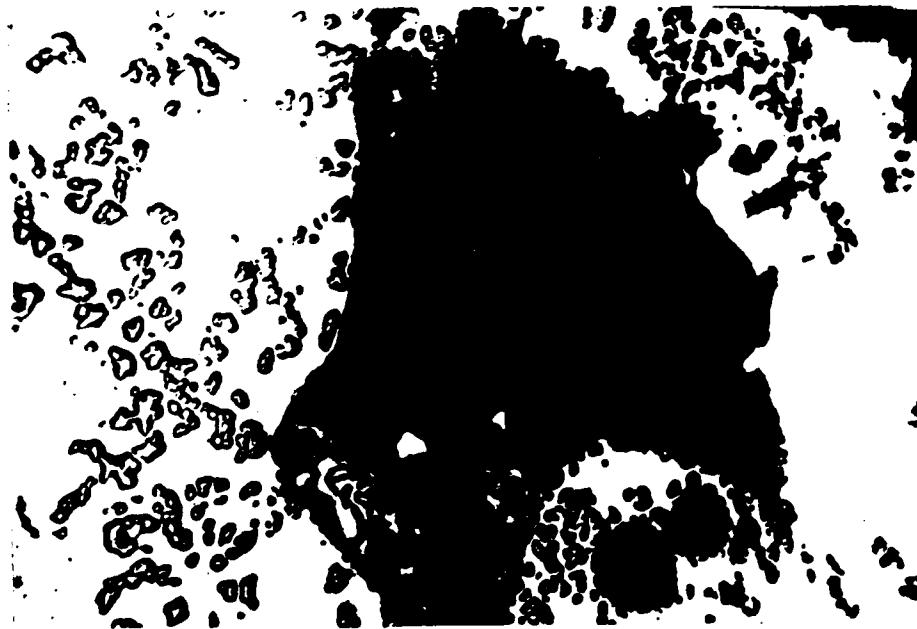


Figure A-13. Small angular inclusions composed of Si, Fe, and U
(dark area is slag, 200X)



Figure A-14. Intermetallic inclusions composed of Cu, Fe, and U,
200X

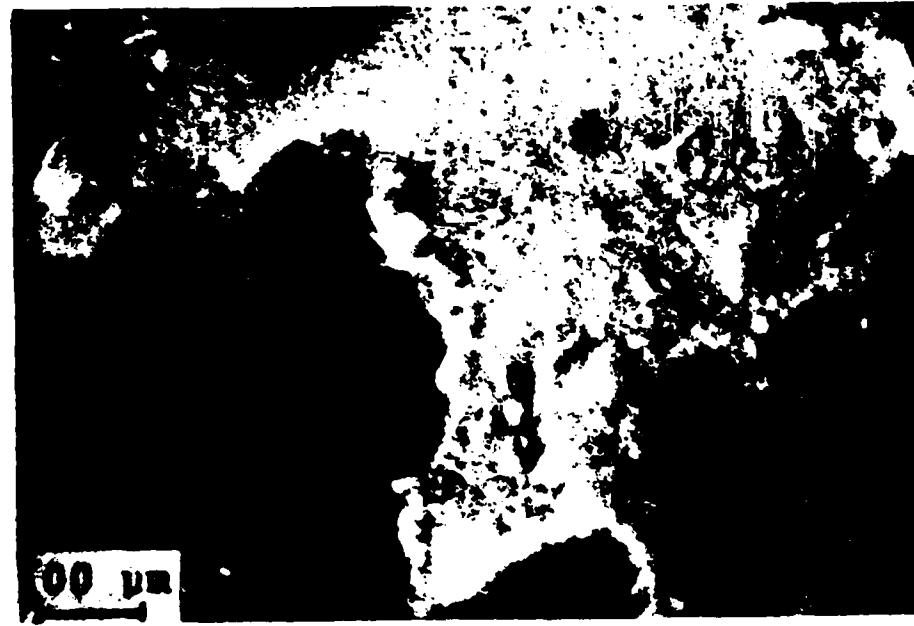


Figure A-15. Slag flux near the top of melt composed of magnesium,
calcium, and silicon (by SEM, 100X)

segregate and form as carbides during remelting of the chips.

5. Slag flux exhibits magnesium, calcium, silicon and titanium. (Figure A-15).

6. All of the grain boundaries contain magnesium. Evidently, they are rich in magnesium fluoride as well as uranium dioxide. (Figure A-16).

7. Dendritic inclusions (Figure A-17) suggest that only the two elements uranium and titanium coexist. The likely identity of the inclusions is an intermetallic phase formed during slow cooling.

X-Ray Diffraction

X-ray diffraction analysis was made specifically for a uranium chip, a uranium rod (U-0.75% Ti), uranium dioxide taken from the sediments of the container of chips, commercial uranium dioxide and some melting products.

1. A piece of a uranium chip suitable for x-ray diffraction analysis was pickled in 5% sulphuric acid (to remove the oxide) and centered appropriately in the x-ray beam. The result confirmed that the chip was pure uranium and no traces of titanium could be observed.

2. The sediment from the bottom of the container of the chips was rinsed with methylene chloride (CH_2CL_2) to remove greases and oils, then filtered and dried in air. Further, the sediment was ground into a powder suitable for powder x-ray diffraction analysis. The diffraction patterns of this sample compared with the patterns of commercial uranium dioxide showed that the sediment was composed primarily of uranium dioxide (fluorite structure). No x-ray evidence of uranium trioxide was detected.

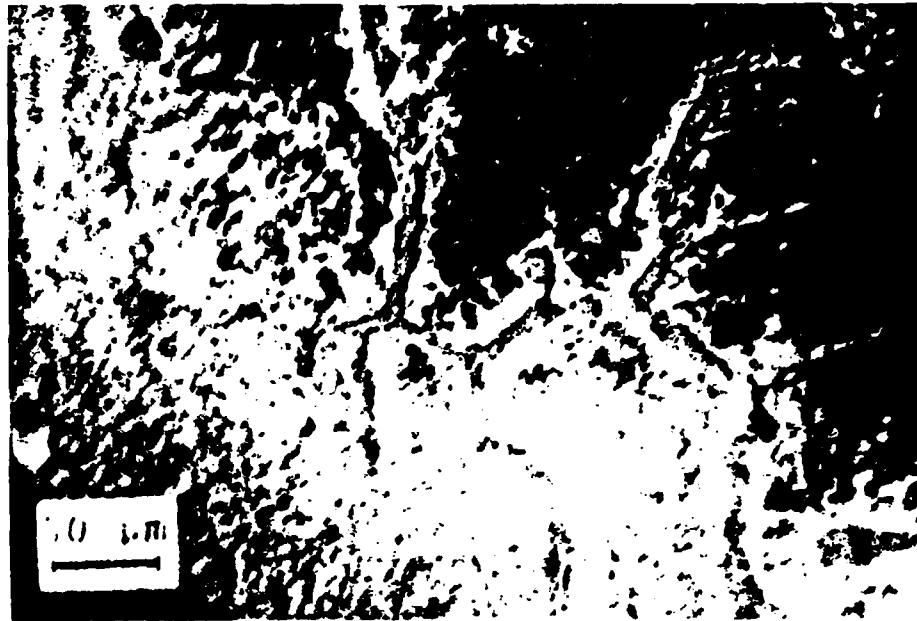


Figure A-16. Grain boundaries composed of magnesium and uranium
(bright field, 200X)



Figure A-17. Dendritic titanium rich inclusions composed of Ti and U
(bright field, 200X)

3. A small disc sample cut from uranium-0.75% titanium rod showed that this metal is alpha uranium (orthorhombic).

4. Several samples obtained by induction melting process were analyzed by the diffraction data available in the card index (16). Most of the patterns were made by pure uranium, a few small patterns were identified as magnesium fluoride and uranium dioxide. No uranium carbide (UC) could be detected.

EXPERIMENTAL RESULTS

After the melt had cooled, the recycled metals were cleaned of salts and weighed to determine yield by comparing with the weight of the chip prior to melting.

Metallographic analysis was next, including Optical Microscope, X-ray Diffractometer, and Energy-Dispersive X-ray (EDX). Chemical analyses were accomplished in certain cases (by a commercial laboratory). (See Table A-1).

Crucible

Crucible materials such as BeO, ThO₂, MgO, CaO and ZrO₂ have been successfully used (1). However, based on economy, graphite, which doubles as an energy coupler for the induction furnace, was used in this study. Its thermal characteristics are such that it may be heated or cooled rapidly and it possesses good strength and shock resistance. Also, graphite is easily machined to a high degree of uniformity. To avoid carbonization, various metal oxide coatings (MgO, BeO, BeSO₄, ZrO₂-SiO₂ and MgZrO₃) were applied by brushing or by slurry techniques.

Table A-1

Specimen number	Chip weight (grams)	Recycled metal weight (grams)	Percent yield	Salt used	Chemical analysis
1	97	22	22.7	MgF ₂	Ti: 0.50% C: 0.012% O: 46 ppm
2	126	30.5	24.2	MgF ₂	Ti: 0.07% C: 0.41% O: 32 ppm
3	190	125	65.8	MgF ₂	Ti: 0.57% C: 0.18% O: 22 ppm
4 (Inert gas system)	114	97	85	MgF ₂	Ti: 0.43% C: 0.195% O: 0.153%
5	165	142	86.1	MgF ₂	Ti: 1.52% C: 0.52% O: 0.473%
6	103	92	89.3	BaCl ₂	Ti: 0.88% C: 0.64% O: 0.215%
7	169	150	88.8	BaCl ₂	Ti: 0.07% C: 0.145% O: 0.37%
8	103	82	79.6	CaCl ₂	Ti: 1.52% C: 0.52% O: 473%

Table A-1 (continued)

Specimen number	Chip weight (grams)	Recycled metal weight (grams)	Percent yield	Salt used	Chemical analysis
9	151	132	87.5	MgF ₂	Ti: 0.03% C: 0.033% O: 0.004%
10	191	168	87.9	MgF ₂	Ti: 0.15% C: 0.102% O: 0.004%

The salt was usually in powder form, except for specimens 9 and 10. In these cases, MgF₂ in bulk (rocky) form was used (a by-product from Mg reduction of UF₄).

Adequate protection from carbon contamination was assured in this way up to approximately 1300°C. Above this temperature, the coating began to spall off and became ineffective. Recently, experimental work with multiple plasma-sprayed layers of niobium and zirconia, molybdenum and zirconia, or niobium, zirconia and yttria have proven to be very effective in reducing carbon contamination at temperatures up to 1550°C. However, these coatings are expensive and somewhat difficult to apply and require specialized equipment. Fortunately, in this research, only small amounts of chips (less than 200 grams) were melted and the molten metal was surrounded by molten salt which minimized carbon contamination. Therefore, none of the various coating procedures were used in this study. The exterior of the crucible was configured in a cone shape. Narrowest at the bottom (10 degrees taper), this shape encouraged coalescing of the molten metal globules. Also, it is believed that the taper (25 to 30 degrees from the horizontal) eased removal of the metal product from the crucible. In order to further facilitate removal of the consolidated metal, a hole was drilled in the bottom (see Appendix C).

To reduce heat loss to the cooling water circulating in the induction coil, a refractory wall of magnesium oxide and asbestos was built between the crucible and the coil. This required some careful adjustment so that adequate electrical coupling between the coil and the crucible was maintained. For the safety of raising the temperature to 1350°C easily, a 1/4 inch thick graphite felt was added between the crucible and the refractory wall in this study and the results have been proved satisfactory.

The Oxidation of Uranium

1. Oxidation of as-cast metal:

Uranium tarnishes rapidly at room temperature. The oxides formed are dark blue or black for the more extensive oxidation and gold or brown for the slight oxidation.

It has been shown that the oxidation starts parabolically and eventually becomes linear (7). A film-cracking hypothesis thus was proposed to explain this transition phenomenon. The volume of oxide formed is thought larger than that of metal consumed; that is, the oxide is under compressive strain. If the strain is too large, the oxide cracks or flakes at a certain thickness, resulting in linear oxidation kinetics.

From the metallographic observation, inclusions may also promote the cracking because of the strain field between the inclusions and metal itself.

Figures A-18, A-19, A-20, A-21 show the oxide films formed on the surface of the as-cast metal.

2. Oxidation of the chips:

- a. The as-received chips, formed by mechanical machining, were heavily oxidized (Figure A-22).

As Figure A-23 shows, the thickness of the oxide layer was about 1/20 of that of the metal. This is not critical and depends on the extent of the chips formed during manufacturing.

- b. Using an X-ray diffractometer to analyse the composition of the oxides on the chips:

The sediment from the bottom of the container of the chips was rinsed with methylene chloride to remove greases and oils, then filtered and dried in air. The sediment was ground into a powder suitable for powder x-ray analysis.

The diffraction patterns of this sample, when compared with the pattern of commercial uranium dioxide showed the sediment to be

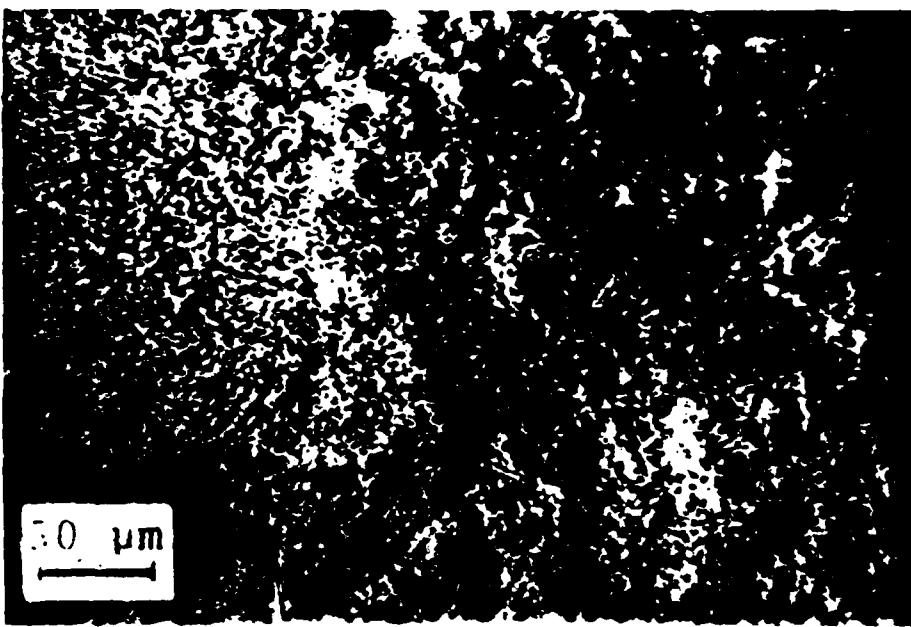


Figure A-18. Oxidation of the polished metal surface after exposure in air six hours (bright field, 200X)



Figure A-19. Oxide films formed and cracked after exposure to air three days (by SEM, 300X)



Figure A-20. Same as Figure 19. (by SEM, 600X)



Figure A-21. Oxide film flakes after exposure to air one week
(by SEM, 300X)



Figure A-22. Oxide layers formed on the machined chips
(by SEM, 5000X)

composed of uranium dioxide which has a fluorite structure. There is no x-ray evidence for the presence of uranium trioxide.

3. Uranium Dioxide UO_2 .

- a. Uranium dioxide has the fluorite structure, $a = 4.4581 \text{ \AA}$, in which uranium atoms are at face-center positions; the oxygen atoms occupy the tetrahedral sites (see Figure A-23).

Alberman and Anderson (9) found that the dioxide will take up oxygen at temperatures below 230°C to a composition UO_{22} without change of structure or appreciable change of cell dimension. Oxides in this composition range decompose, when annealed at high temperature, into well-defined cubic phases, viz, UO_2 and a phase of composition close up to UO_{22} (" UO_2 phase").

Oxides in the composition range $\text{UO}_{22} - \text{UO}_{23}$ prepared below 230°C have a tetragonal structure with axial ratio c/a close to unity and change progressively with composition; these oxides decompose on heating into the β -cubic phase and a U_3O_8 - like phase. The relation between cell dimensions and composition is shown graphically in Figure A-25.

The oxidation of UO_2 at low temperature thus forms, successfully, two closely related non-stoichiometric phases: the cubic oxides $\text{UO}_{20} - \text{UO}_{22}$ and the tetragonal oxides $\text{UO}_{22} - \text{UO}_{23}$.

Both of these are to be considered as defect structure, containing a stoichiometric excess of oxygen in the interstitial positions, and the cation lattice is substantially perfect.

b. The Oxidation of UO_2 :

It has been reported by Alberman and Anderson (9) that the oxidation of UO_2 is characterized by an extremely rapid stage, declining

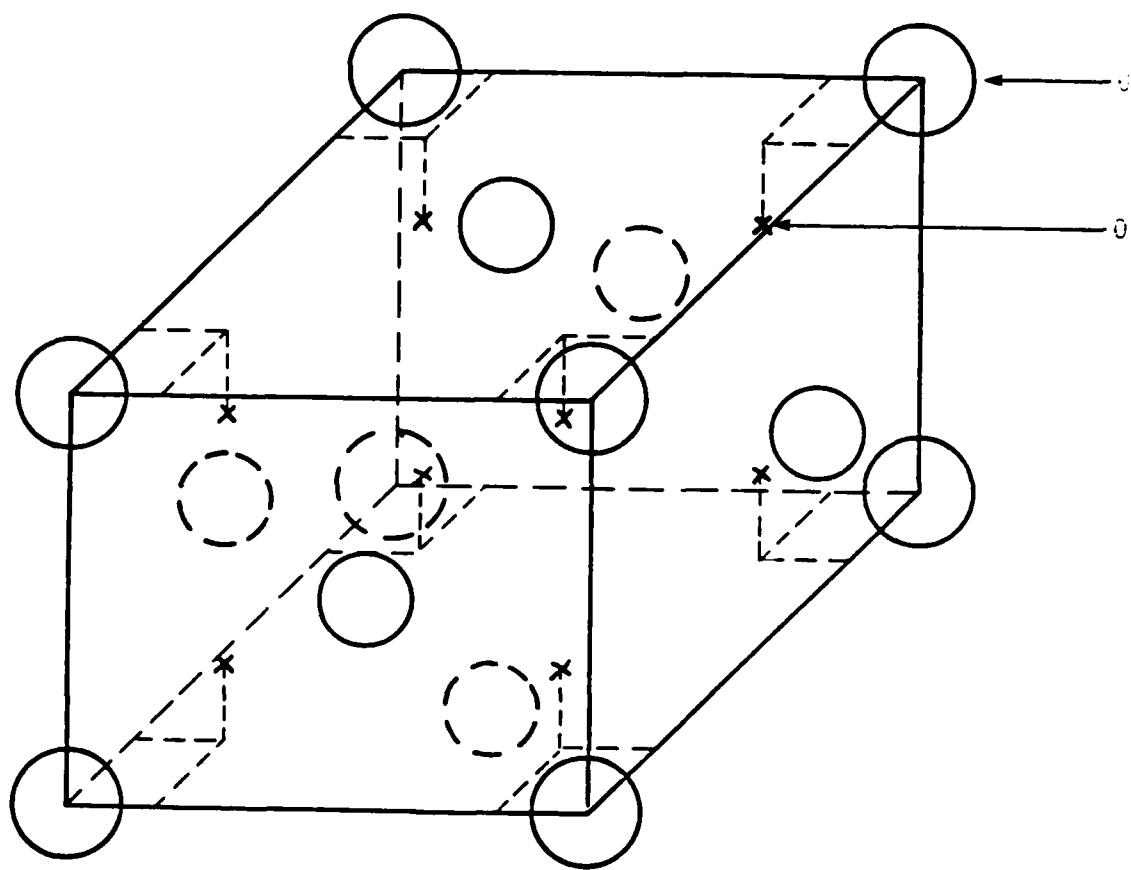


Figure A-23. The Structure of UO_2

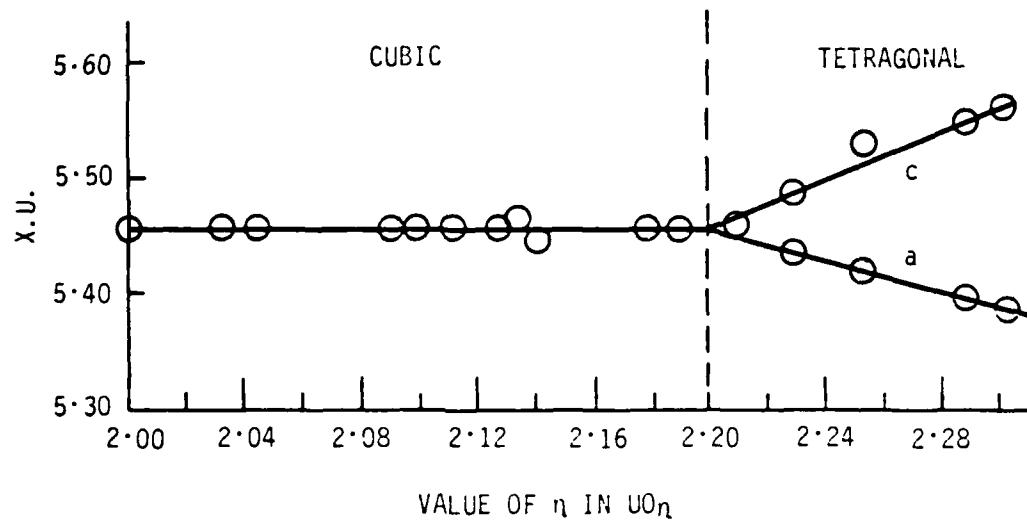


Figure A-24. The relation between cell dimensions and composition of UO_2

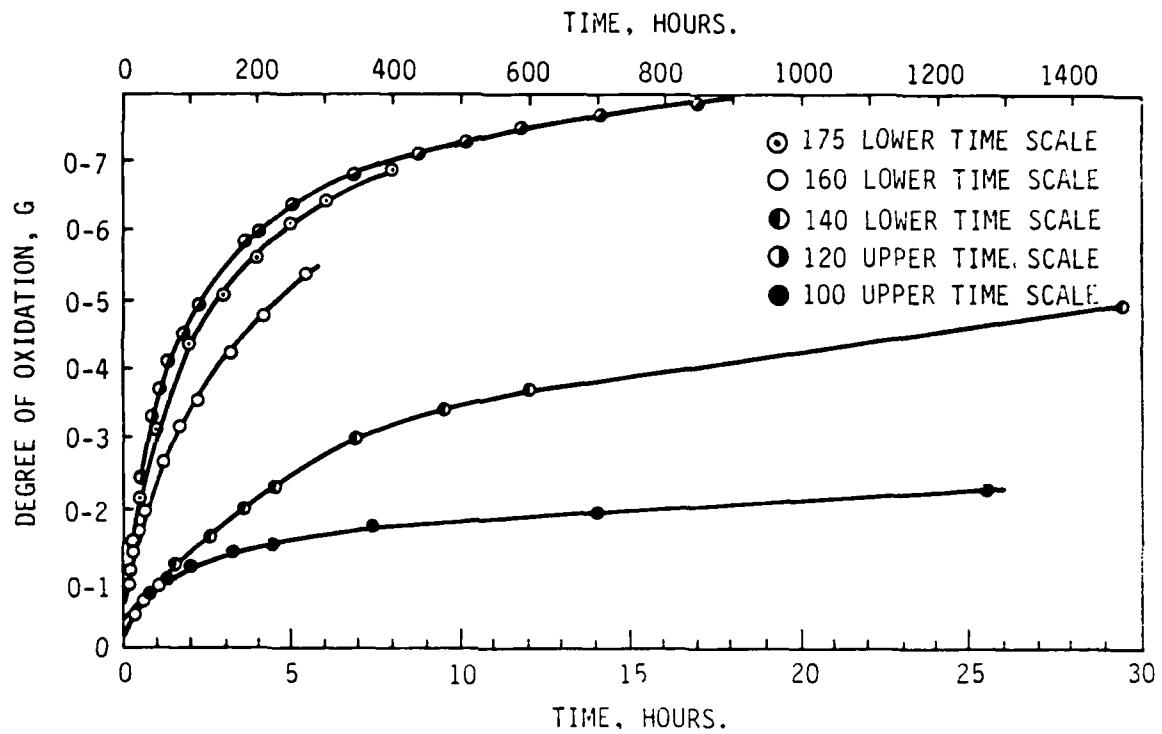


FIGURE 25. OXIDATION OF UO_2 AT VARIOUS TEMPERATURES.
 (CITED FORM THE SAME JOURNAL AS FIGURE 25).

Figure A-25. Oxidation of UO_2 at various temperatures

regularly according to a roughly parabolic law which suggests that the reaction is a typical diffusion-controlled process. The reaction can be expressed as:



In the case for the oxidation of UO_2 , the solid compound A has a wide range of composition (UO_{20} - UO_{25}). So the reaction does not produce a new phase (as long as the crystal structure remains unchanged); rather a composition gradient will be established through each grain as reaction proceeds. The distribution of composition is similar to the distribution of temperature in a spherical particle immersed in a heated environment; the kinetics of the reactions are determined by the diffusion of reactant through the particle and the appropriate solution of the diffusion equation can be taken from the analogous case of the conduction of heat.

$$\frac{x_t - x_i}{x_f - x_i} = F; \text{ the fraction of conversion (a variable)}$$

Where x_i : the initial composition

x_t : the composition at time t.

x_f : ultimate composition

Thermodynamic Considerations in Choosing the Salt

Many difficulties which associate with the recycling of the chips come from the high reactivity of uranium with atmospheric oxygen resulting in the formation of a highly stable uranium dioxide.

The most common and severe phenomenon in the "melting processes" is the continuous oxidation of the chips. But the addition of molten salt greatly reduces the oxidation by acting as a flux and dissolving or

dispersing the uranium oxides which then float and become slags. Meanwhile, the molten metal flows downward and coalesces.

Magnesium fluoride, calcium chloride, and barium chloride have been tried and proved to be effective.

Since the interstices which exist in the salt and chips still contain some atmospheric gas, it is preferable to liquify the salt before the uranium chips are introduced.

Usually, the molten salt appears to boil because of the electromagnetic stirring from the induction coil. This is beneficial for the removal of the coated oxides by the fluxing molten salt; also, it is helpful for coalescing of the melt. But the liquid salt depth should be at least one inch above the melting chips to minimize exposure of the chips to oxidizing gases at the surface.

As mentioned before, the salts only act as a flux and dissolve the oxides. It is possible that the addition of reducing agents to the molten salt will react with uranium oxides by the processes:

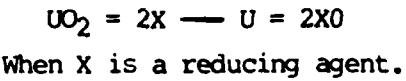


Figure A-26 (3) shows the free energies of formation of uranium dioxide, calcium oxide and magnesium oxide plotted as functions of temperature. From Figure A-26, it is evident that calcium will reduce uranium dioxide to form uranium metal at any temperature in the normal operating range whereas magnesium will reduce uranium oxide only at temperatures below about 1270°C; slightly above this temperature the reverse reaction occurs.

Further, since magnesium boils at 1117°C, operating above this temperature causes high pressures of magnesium vapor to be developed, thus

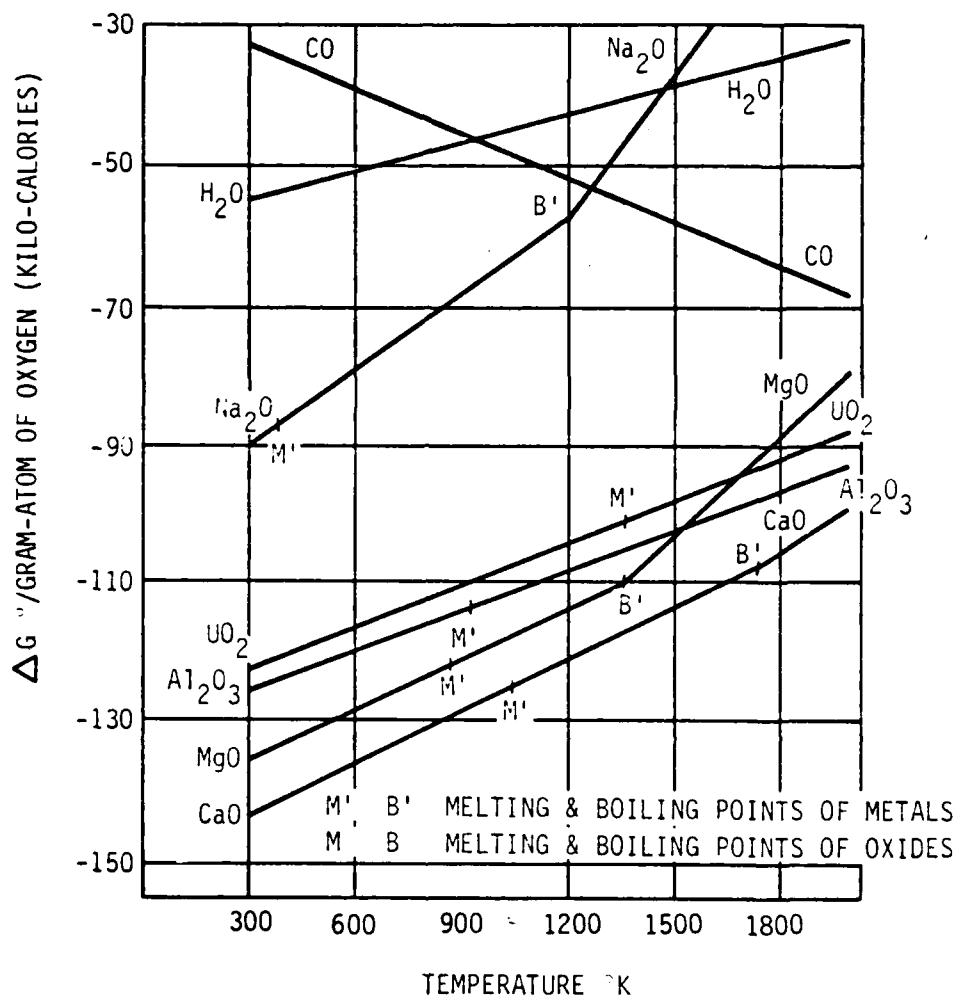


Figure A-26. Free energies of formation of oxides as a function of temperature

magnesium was ruled out in favor of calcium metal.

The heat of reaction of calcium with UO_2 at 298°K is 45 kilocalories per gram-atom of uranium (3). It would seem then, that if the chips were dropped at the melting point of uranium, the heat of reaction from the Ca, UO_2 reaction would help to sustain the molten state, thus improving the melting of the chips and possibly reducing the holding time at high temperature (thereby diminishing carbon pick-up from the graphite crucible).

The amount of calcium necessary is determined by the amount of oxide present on the chips. In this study, about 2 grams of calcium metal were added to 100 grams of uranium chips. The excess amount of calcium with the lime formed in the reaction would dissolve into molten salt and separate with the uranium metal. In order to facilitate uniform reactions with the oxides, the calcium metal was comminuted and mixed completely with the powdered salt which then acted as both a flux and a reactant in the presence of chips. It is important to note here that the calcium metal, like the magnesium metal, tends to distill from the reaction mass and, once this occurs, it reacts violently. Fortunately, the melt product showed that large quantities of oxide dropped to the top of the ingot without adding calcium metal. Therefore, the density differential cleansing of oxides is favored over the reduction of UO by calcium metal. That is, the mechanical reduction of oxides is the most significant benefit when the salt is fluxing. Consequently, all subsequent melting tests were performed using a single salt, either MgF_2 or BaCl_2 .

It was thus concluded that the reduction of UO by Ca, though thermodynamically feasible, was not practical. The principal object was to insure that the uranium and the slag become sufficiently fluid. The salt should have a melting temperature lower than the metal (to maintain a continuous protective slag envelope).

Magnesium fluoride (M.P. = 1261°C, B.P. = 2239°C), including magnesium fluoride generated in the reduction of UF by magnesium metal, was found to be the most satisfactory salt. Barium chloride (M.P. = 963°C, B.P. = 1560°C) was also satisfactory. Calcium chloride, however, is too volatile and deliquescent.

The best operational procedure involved premelting the salt in the crucible and then charging the pellets into the molten salt at 1450°C. The temperature dropped immediately to about 1250 - 1300°C, depending on the amount of chips that were added. The chips should be fully immersed in the molten salt. Experience showed that holding the molten bath of metal and salt at 1350°C about 30 minutes resulted in the best metallic product.

SUMMARY OF CONCLUSIONS

Melting Process

1. Good atmosphere control is definitely necessary. A preliminary outgassing treatment of the crucible and charging slags is advisable. A furnace pressure of approximately a half atmosphere (15 inches of mercury) with a backfill of an inert gas was found to be satisfactory.
2. It is advantageous to melt at the lowest temperature and shortest hold time possible to prevent contamination of carbon from the crucible. However, temperatures that are too low do not allow adequate mixing between the slag and molten metal. The most appropriate temperature is 1350°C for about 30 minutes.
3. Eutectic calcium fluoride - magnesium fluoride composition (for example, 52 weight percent of CaF₂ - 48 percent of MgF₂) was used unsuccessfully. 100 percent MgF₂ was most acceptable. Pure BaCl₂ was also effective..
4. Slags must melt at a lower temperature than the metal. Complete slag coverage of the molten metal was essential since the slag not only provided for refining the metal but also insulated the molten pool from the atmosphere.

Metallography

1. Metallographic and EDX data confirmed that slag entrapment and inclusions were present only at the top of the as-cast metal.
2. Segregation resulting from the melting process is chiefly due to carbide formation of the alloying ingredients. This segregation is

believed to substantially reduce the titanium content of the alloy since the titanium, tied up as a carbide, floats up near the top of the melt.

3. Most of the alloying intermetallic phases appeared as dendrites, especially the titanium rich phase. This is because the addition of titanium above the eutectic composition, increases the liquids temperature. During solidification, as dendritic growth occurs, titanium rich solid is first to form, thus depleting the titanium content of the adjacent liquid. This difference in concentration is further aggravated by the large density difference between the alloying elements and uranium. Thus, most of the segregations rich in alloying elements are found only near the upper part of the melt product.

APPENDIX B
SPECIMEN PREPARATION

As-Received Form of the Chips

No specific size or shape; submerged in mineral oil in metal drums.
The oxidation of the chips ranged from slight to severe.

Preparation for Melting

1. Cleaned by CH_2CL_2 to remove greases and oils.
2. Dried in air.
3. Compressed in cylindrical die (diameter: 3 inches, thickness: 1/2 inch at 4,200 psi for 5 minutes.
4. Coated with salt (MgF_2) to retard oxidation during heat-up in the crucible.
5. Several stages of chip preparation are shown in Figures B-1, B-2, B-3.



Figure B-1. As-Received form of the chips



Figure B-2. Compressed pellets of the chips

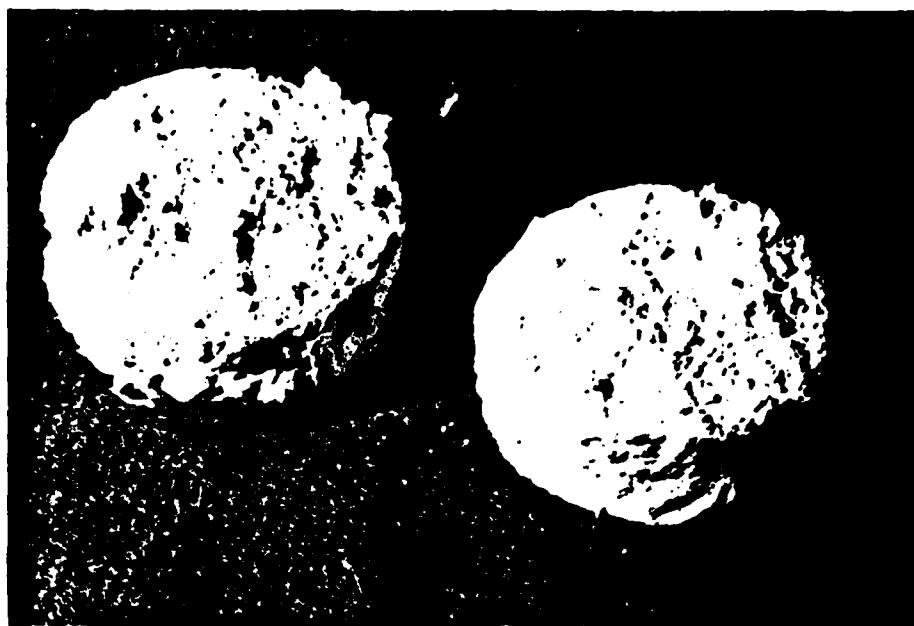


Figure B-3. Chips coated with salts

APPENDIX C
CRUCIBLE

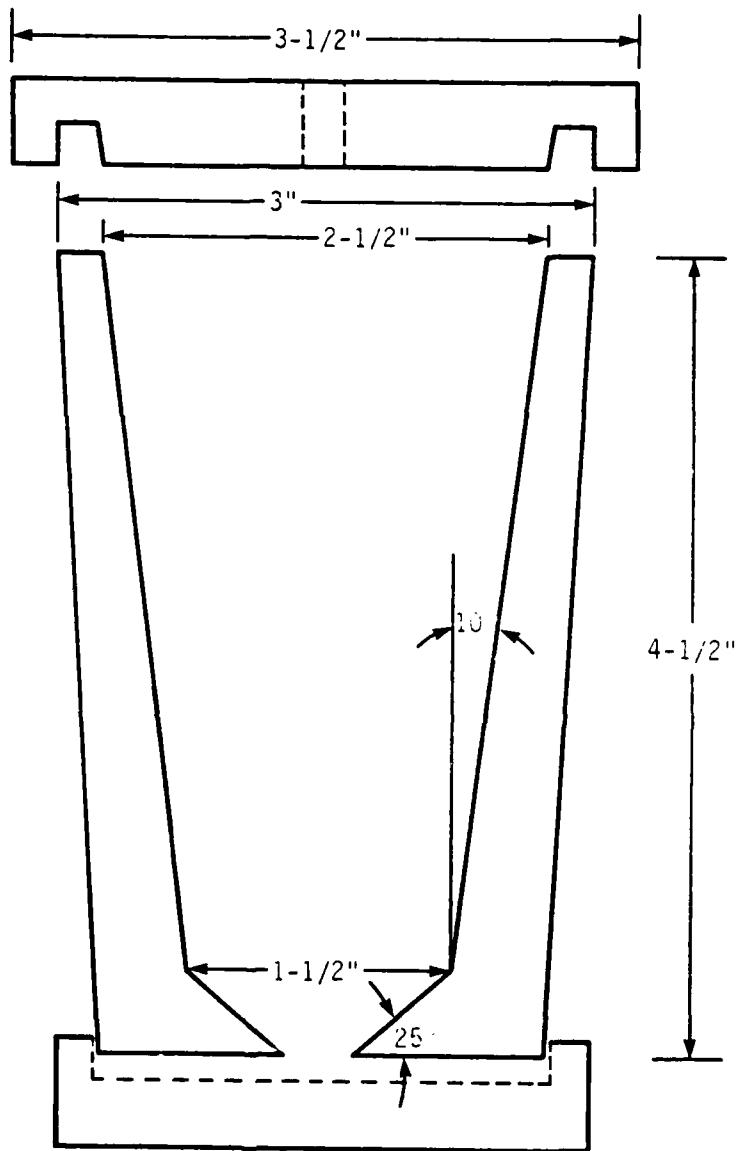


Figure C-1. Crucible

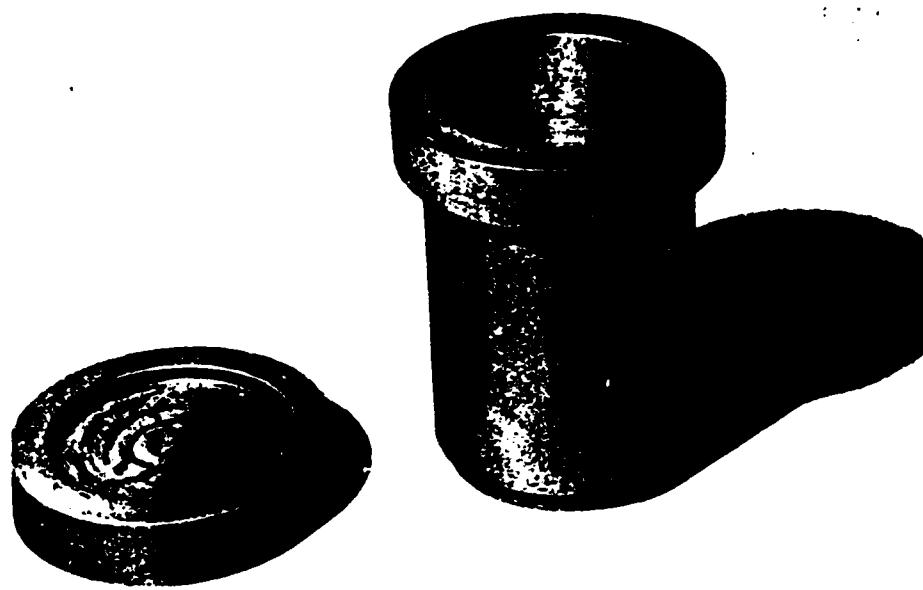


Figure C-2

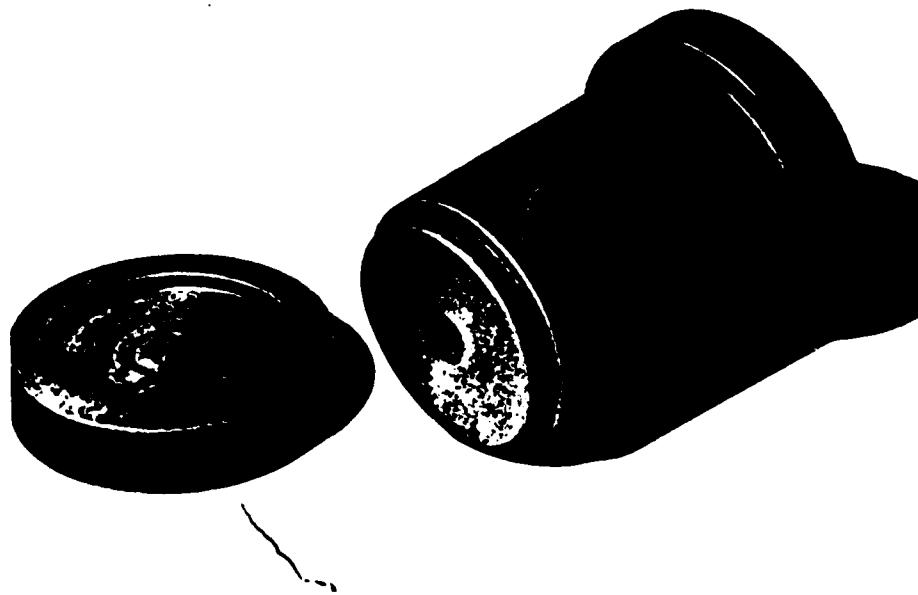


Figure C-3

ADDENDIX D
FURNACE DESCRIPTION

Fire Brick Furnace

A cubic box (15" x 15" x 15"), made of K-30 fire bricks was constructed around an appropriate induction coil. An argon inlet was placed above the crucible. Access was gained by a removable cover (see schematics diagram, Figure D-1).

The charging of reactants or the measuring of temperature were performed directly through the top hole.

Vacuum Induction Melting Furnace

In order to minimize heat loss from the furnace, a double-walled tank furnace was constructed. Aluminum plates were used for the outer wall and asbestos plates were used for the inner wall (Figure D-2).

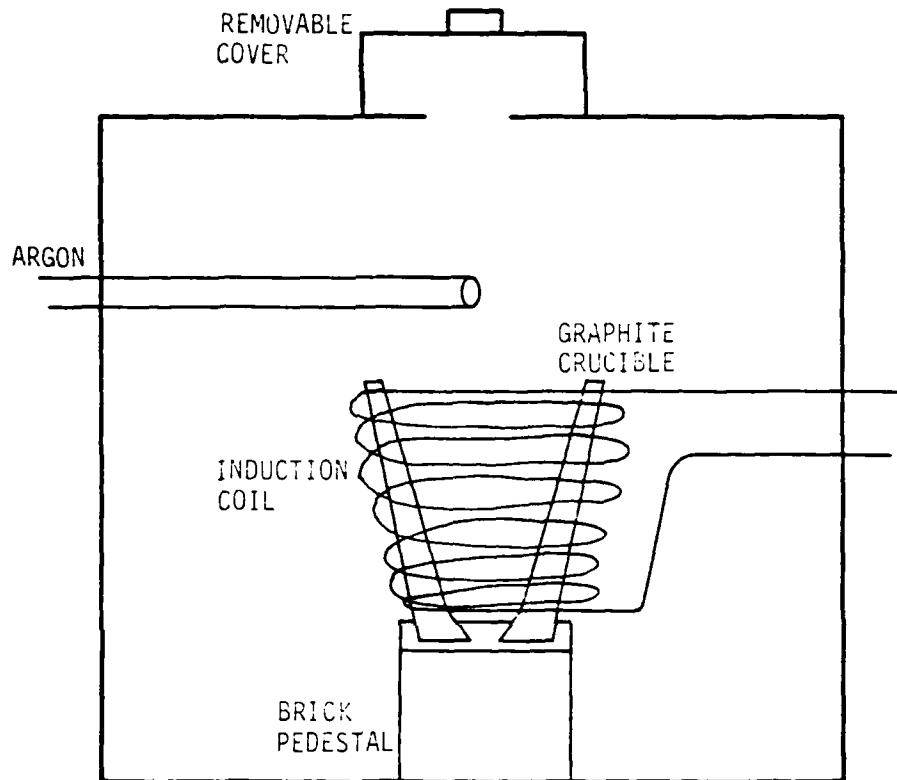


Figure D-1. Brick wall furnace

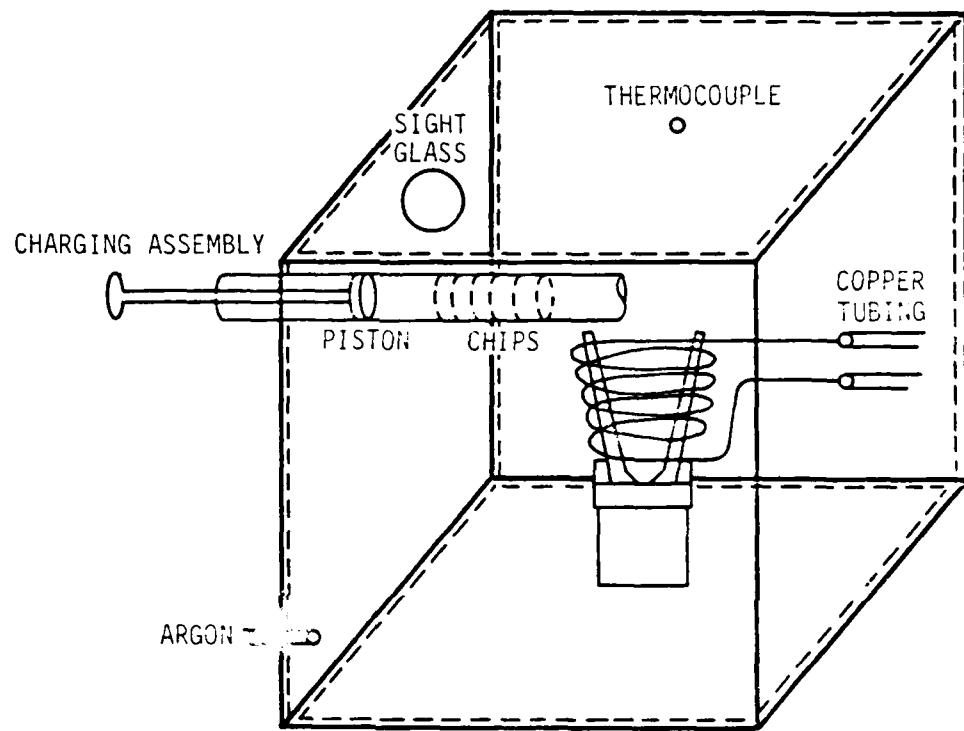


Figure D-2. Vacuum induction furnace

APPENDIX E
ELECTRICAL EQUIPMENT

Generator

The induction heating unit was manufactured by TEK SPECIALTIES, INC. and donated to WPI by Nuclear Metals Incorporated.

Frequency 10 KHZ

Maximum Power 20 KW

Coil

The coil which is made of copper tube (3/8 inch diameter) was wound to fit the crucible shape. Cooling water is circulating in the coil to prevent thermal damage. The pressure of the water was 20 psi all through the melting operation.

Care must be taken to guard against water leakage. The reaction between water and the molten metal could be explosive. Thus inspections were made periodically.

APPENDIX F
TIME, TEMPERATURE, AND OPERATIONS DURING MELTING

Time (min,sec)	Temperature (°C)	Kilowatts	Operations
0	25	40	
2: 10	100		
3: 30	150		
4: 10	200		
4: 50	250		
5: 15	300		
5: 45	350		



Time (min,sec)	Temperature (°C)	Kilowatts	Operations
6: 20	400	40	
6: 55	450		
7: 25	500		
8: 00	550		
8: 35	600		
9: 10	650		
9: 50	700		↓
10: 30	750	60	
11: 10	800		
11: 50	850		
12: 25	900		
13: 12	950		
13: 25	1000		
13: 35	1050		
13: 55	1200		
14: 10	1250		↓

Time (min, sec)	Temperature (°C)	Kilowatts	Operations
15: 15	1300	80	
15: 45	1350		
16: 15	1400	90	5. Drop the chips, followed by dropping the salts into crucible from the glass tube.
17: 00	1150		
17: 25	1200		
17: 45	1250		
18: 05	1300		
18: 30	1350		
18: 50	1400		
19: 15	1450		
30: 00	1455	80	
31: 40	1400		
56: 05	1380	75	
57: 00	1365		
63: 00	1365	0	6. Shut off the machine.

APPENDIX G
METALLOGRAPHIC PREPARATION

Cutting, Mounting, Polishing and Etching of the Uranium Metal

Cutting

Because of the pyrophoric nature of the uranium, high heat is created when it is cut. The danger of fire as well as the possibility of uranium contaminated fumes are ever present. Therefore, care should be taken in all processing. Enough water applied via hose lines using fog stream for maximum cooling is definitely necessary. Also, a shade to surround the cutting set and protect the operator from being hurt by the flying chips is necessary too.

Those scraps left on the bottom of container have to be wiped and rinsed completely, collected together and put into a special tank for proper disposal.

Mounting

Standard metallographic mounting procedures were used.

Polishing

A series of silicon carbide grits are used for grinding the metal:
NO. 180, 220, 320, 400 and 600.

During each step of grinding, enough water is applied to cool down the metal. Meanwhile, glycerol is added as lubricant. The final grinding on worn 600-grit paper should be performed carefully to keep the scratches as fine as possible.

After washing and drying, the specimen is polished on a wheel covered with a nap cloth. Aluminum oxide (Al_2O_3) with particles size 1 microns, 0.3 microns and 0.05 microns are used as abrasive. The specimen is then washed thoroughly in alcohol and dried by hair dryer.

Usually, a satisfactory surface can be obtained from the above processes, but the best result is accomplished by electrolytic polishing. The phosphoric acid-alcohol bath, composed of 5 parts of phosphoric acid, 5 parts ethylene glycol and 8 parts alcohol used at a current density between 10 and 30 amp/cm² were used in this study.

Etching

Two kinds of etching agents are recommended (14):

1. HNO_3 _____ 38 ml.
HF _____ 1 ml.
Distilled water _____ 100 ml.

2. HNO_3 _____ 30 ml.
Glacial acetic _____ 30 ml.
Glycerol _____ 30 ml.

Experiences show that the first kind is more effective and the most appropriate time is about 80 to 90 seconds. After etching, the sample is cleaned by distilled water and then dried quickly.

APPENDIX H
PRECAUTIONS IN RECYCLING DEPLETED URANIUM

Recognize the Depleted Uranium

Depleted uranium is the product of processing naturally occurring uranium to separate the radioactive U-235. The resultant nearly pure U-238 is only mildly radioactive.

Handling precautions are specified by the Nuclear Regulatory Commission. In addition to the radioactivity hazard, however, there are two others. a). The toxicity of the substance owing to its high atomic number, and b). The fire hazard owing to its pyrophoric nature (a propensity to oxidize).

Radiation exposure to the body by depleted uranium is usually not a problem as its low radiation emissions would generate a significant dose only after an exposure to large quantities of the metal for extended periods of time. The permissible limits for radiation exposure are equivalent to an average of 100 mrem of exposure to whole body radiation per week and 1500 mrem to the hands or feet. A dose of 1500 m/rem to the hands would require a person to physically hold a piece of depleted uranium for a period of approximately 6 full hours per week which is most unlikely except for hand polishing operations. Experience also indicates that persons working with depleted uranium have such a limited exposure to radiation that the use of a film badge or dosimeter is usually not necessary. Also the keeping of personal monitoring records is waived.

These comments are general in nature as it is possible under extreme circumstances to receive a dose requiring personal monitoring and record keeping. However, to do this a person would probably have to be in an area of massive quantities of the metal (tons) and remain in very close proximity to it (2 or 3 feet) for 8 hours per day, or be exposed to depleted uranium daughter products during melting. Experience has shown

that operations such as machining, weighing, grinding, shipping, handling and storing of depleted uranium do not subject a person to significant radiation exposure.

Inhalation or Ingestion of Depleted Uranium Particles

This material, unlike most radioactive materials, emits 3 types of radiation — alpha, beta and gamma. The alpha radiation, which is insignificant as an external radiation hazard, becomes a problem once uranium enters the body by inhalation or ingestion. Once in the body, alpha rays, because of their short range and high ionization, are potentially more hazardous than beta or gamma radiation. However, over the time period since the Manhattan Project, the Atomic Energy Commission and Nuclear Regulatory Commission have been without incident toxicologically due to depleted uranium. Persons using depleted uranium in such a manner as to create dust, mist, or fumes are created by grinding, turning, melting, welding, abrading, hot-rolling, extrusion, forging, etc.

Experience has shown that in order to conform to the regulations, the following procedures are necessary; a.) proper ventilation must be installed to conform with the operation. b.) If the operation is one in which the depleted uranium is processed 8 hours per day for a long period of time (12 months or more) filters must be required on the discharge side of the vent system. c.) In order to show that such air in and outside of the plant conforms to permissible limits, proper monitoring procedures are required.

Ingestion or Eating of Depleted Uranium is a Hazard

A person working with lead or steel refrains from eating the material accidentally by washing his hands before eating. The same applies to the handling of uranium. Normal personal hygiene procedures are all that is required to control this hazard.

The Pyrophoricity

Uranium metal in depleted, normal or enriched form will burn under certain circumstances and in rare cases explode with considerable violence.

During turning or grinding of the metal, chips or turnings are created which pose a fire hazard if not handled properly. Normally such chips are collected from under the chuck of a lathe when the quantity reaches about a handful. The chips are then put into a covered pail or receptacle usually not in excess of 30 gallons in size. A layer of used motor oil or water in the container is always kept about 2 inches over the surface of the stored chips. This material keeps the chips from spontaneous heating and taking fire while in storage.

Having only a limited amount of scrap material at the point of operation under the chuck or wheel makes a large or dangerous fire impossible. Usually such fires are extinguished by dispersing or scattering the chips about the lathe bed carefully to dissipate the heat and reduce the chips to below ignition temperature. The exhaust ventilation in such areas of operation is sufficient enough to vent fumes from a small fire involving a handful of chips.

In order to assure the safety, the following procedures are usually involved: a.) Completely submerge the burning metal in a large container of water. b.) Use proper fire extinguishers such as Ansul Met-L-X directly

on the burning metal. c.) Water applied via hose lines using fog stream for maximum cooling.

Waste Disposal

Unusual scrap-wipers-rags-sludges from machines, etc. and general wastes contaminated with depleted uranium cannot be disposed of by the usual disposal procedures. Disposing of such material in a public dump is not permissible. Any waste must be handled according to regulations of the Nuclear Regulatory Commission.

Personal Protective Equipment

- a. For the handling of depleted uranium: gloves.
- b. For the melting of depleted uranium: gloves, respirator.
- c. For the cutting and polishing of depleted uranium: gloves, respirator, coveralls.

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APPENDIX I
SPECIFICATION FOR
ELECTRON-BEAM VACUUM MELTING AND CASTING FURNACE

Specification Number NM-3800 A

Prepared By

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Nuclear Metals, Inc.

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September 1982

1.0 SCOPE

1.1 The term "equipment", as used herein, shall be construed to mean the basic item identified by the title of this specification and described in paragraphs 1.2 and 4.2.1. The equipment is the complete unit with all systems tooling and other auxiliary items required to meet the requirements defined herein.

1.2 This specification defines the requirements of an Electron-Beam Furnace to melt machine chips of a depleted uranium-titanium alloy and to produce ingots of a size suitable for vacuum induction remelting and casting.

2.0 REFERENCE DOCUMENTS

2.1 The system to be furnished under this specification shall comply with the following standards of date and issue in effect on the date of this proposal. Should the Vendor find any conflict between these standards, he shall immediately contact Nuclear Metals, Inc. who shall determine the order of precedence.

2.1.1 Military Standards

2.1.1.1 MIL-STD-1188A Supplies and Equipment.

Commercial Packaging

MIL-H-19457B (Ships) Type 1.

2.1.2 Other Publications

2.1.2.1 Occupational Safety and Health Act, Public Law 91-956, 91st Congress 52193, 29 December 1970.

2.1.2.2 Joint Industrial Council

2.1.2.2.1 Electrical Standards for Mass Production Equipment
EMP-1-67 and General Purpose Machine Tools EGP-1-67.

2.1.2.2.2 Pneumatic Standards for Industrial Equipment P-1.

2.1.2.2.3 Hydraulic Standards for Industrial Equipment H-1
Occupational Safety and Health Act, Public Law 91-956, 91st Congress
52193, 29 December 1970.

2.1.2.2.4 American National Standards Institute (ANSI).

2.1.3 Electrical Equipment

2.1.3.1 National Board of Fire Underwriters

2.1.3.2 National Electrical Manufacturing Association (NEMA)

2.1.3.3 Institute of Electrical and Electronic Engineers (IEEE)

2.1.3.4 National Electric Code

2.1.4 Pneumatic System

2.1.4.1 American Society of Mechanical Engineers (ASME), Section
VIII-Standards for Unfired Pressure Vessels

3.0 INPUT AND OUTPUT OF THE OPERATION

3.1 Input Material to Operation

3.1.1 Depleted uranium-titanium alloy machine chips with some
possible solid pieces having a maximum dimension of 1.5 inches by 2 to 4
inches long, free of all cutting oils, solvents and moisture, but having
an oxide coating. Recommendation is requested as to maximum size of solids
that may be considered for feed material.

3.2 Output Material from the Furnace

3.2.1 Cast ingot from a nominal 304mm (12.0 inch) diameter to 342mm (13.5 inch) diameter by 355mm (14.0 inch) to 465mm (18.0 inches) long, weighing a nominal 636 kg (1400 lb).

3.2.2 Design consideration will be given, subject to cost and engineering constraints, to the possible accommodation of a continuous casting mode for the production of 114 mm (4.5 inch) diameter billets of flexible length.

3.3 Throughput of Operation

3.3.1 The required monthly throughput of the equipment is defined as follows:

Units	Level 1	Level 2	Level 3
Lbs/Month	39,200	119,000	239,400
Kg/Month	17,796	54,026	108,687

3.4 Efficiency

3.4.1 Efficiency or up-time is to be considered as 70 percent or, on the basis of a 500 hour month, the equipment will be available to perform its function 350 hours. Should the Vendor believe that a 70 percent efficiency factor is not correct for his equipment, he shall so state in his quotation.

4.0 REQUIREMENTS

4.1 General Requirements

4.1.1 Workmanship shall be of high quality and shall be demonstrated to be of a level consistent with the requirements of this specification.

4.1.1.1 All belt or chain drives, sheaves and pulleys, couplings, gear shaft extensions with projecting set screws, keys, slots or other rotating parts as well as all pinch points and working parts shall be enclosed or adequately guarded so that it will be impossible under normal operating conditions for the operator's person or clothing to contact such moving parts or pinch points.

4.1.1.2 All guards shall be easily removable for maintenance purposes such as inspection, adjustment, lubrication and part replacement.

4.1.1.3 Provisions shall be made to permit adjustment to compensate for wear of components. All parts subject to adjustment or replacement shall be readily accessible.

4.1.1.4 All replacement parts shall be manufactured to definite standards for tolerance, clearance and finish so that they may be field installed without further machining.

4.1.1.5 All equipment to be furnished by the Seller shall be designed to satisfy the safety and occupational noise requirements set forth in Section 2.0 of this specification.

4.1.1.6 Radiation shielding requirements shall be such that at any point outside the furnace during a maximum possible loading and/or at peak operation voltage, the radiation field shall be limited to 2 MR/hour at contact when depleted uranium is being melted.

4.1.2 Electrical Requirements. All electrical equipment and controls shall comply with the standards referenced in Section 2.0. All equipment shall be rated for 110 volt single (1) phase, sixty (60) cycle

power input. All control circuits to be 110 volts.

4.1.3 Hydraulic Systems. All hydraulic systems shall utilize non-flammable fluids.

4.1.4 Pneumatic Systems. All pneumatic systems shall conform to the ASME codes.

4.1.5 Lubrication. Critical points should be automatically lubricated whenever possible.

4.1.6 Lifting Devices. All components weighing fifty (50) pounds or more to be provided with lifting rings, lugs, tapped eye bolt holes or other equally suitable means to facilitate movement and positioning of the equipment with overhead or mobile cranes. The lifting points shall be selected to assure maximum safety to rigging and maintenance personnel, to preclude damage to the equipment, and to provide proper balance when erecting and installing the equipment. All lifting points shall be clearly marked in a manner that will preclude obliteration by paint, or print which clearly shows the lifting points shall be provided. If this is not standard it shall be quoted as an option.

4.1.7 Paint. The equipment shall be finished in the manufacturer's normal method and colors.

4.1.8 Special Health and Safety Requirements. Depleted uranium and its alloys are slightly radioactive (classified as low specific activity) and uranium is a heavy metal poison. Depleted uranium is also pyrophoric. All equipment shall be equipped or capable of being equipped with ventilation wherever vapors of finely divided particles of uranium or uranium oxide are generated. All equipment shall be constructed to contain all processing fluids (coolants, oils, etc.) which come in contact with the depleted uranium alloy during processing. Equipment shall also restrict the expulsion of any finely divided material (chips, oxides, etc.)

from the equipment. Equipment must be constructed to permit the cleaning of residual depleted uranium on a periodic basis.

4.1.9 Layaway. The equipment may be removed from production and placed in layaway for indefinite periods. The Vendor shall consider this possibility as it relates to his equipment. Further quotation requirements concerning layaway are found in Section 5.

4.1.10 Special Tooling or Modifications. Prior to fabrication of the equipment, the Vendor shall provide for Nuclear Metals, Inc. approval, all drawings and related operational procedures describing any special tooling or modification to a standard piece of equipment required to meet the requirements of the specification. Nuclear Metals, Inc. shall review the designs and shall return them with approval or comments within fifteen (15) days after receipt at Nuclear Metals, Inc. Standard equipment not requiring modification does not require approval prior to fabrication.

4.1.11 Performance Guarantee

4.1.11.1 The Vendor shall state in his offer a guarantee of equipment performance. This performance guarantee shall be the basis for acceptance of the equipment by Nuclear Metals, Inc. The Vendor's statement shall be specific regarding cycle times which is defined herein as floor-to-floor time (can be expressed as throughput in weight per hour or month) and whether the equipment will meet the requirements of this specification. Any requirements from which the Vendor deviates or takes exception shall be clearly stated.

4.1.11.2 The depleted uranium alloy used in the fabrication of the item stated herein is regulated by the Federal Government or by State Government. A facility must be licensed by the Federal Government or State Government to receive or process the material. Thus it is not feasible to furnish the material to the Vendor for work in the Vendor's facility. Should a substitute material be considered useful for trials in

a Vendor's plant, it shall be so stated under paragraph 4.2.

4.1.11.3 Prior to delivery, acceptance tests shall be conducted at the Vendor's plant. Since these tests cannot be conducted with the depleted uranium alloy, the acceptance for the delivery shall be based upon a demonstration that the equipment shall:

- a. Perform the function as described by this specification and the Vendor's proposal.
- b. Be capable of meeting the floor-to-floor time quoted by the Vendor.
- c. Provide the accuracy and repeatability required by this specification and appended drawings.

4.1.11.4 Once the equipment is installed at the Nuclear Metals, Inc. facility, a proveout run shall be made by the Vendor. Final acceptance of the equipment by Nuclear Metals, Inc. shall be based upon achievement of the performance guarantee during the proveout trials. The proveout shall consist of two (2) melting and casting cycles in two (2) consecutive shifts.

4.2 Specific Requirements

4.2.1 Basic Furnace Design. The overall furnace design should be such as to allow for the following:

4.2.1.1 A side chamber(s) to serve as a loading port for the feed material having the capability of being evacuated and isolated from the main melt chamber. Mechanical agitation of dry machine turnings in air is extremely hazardous. Friction between one dry chip and another can cause spontaneous ignition.

4.2.1.2 Some mechanism by which material can safely be transferred from the auxiliary to the main chamber.

4.2.1.3 A hearth is required in which the melting would be accomplished. The nature of the feed material having an oxide becomes a major concern for the ultimate quality of the cast ingot. Some means will need to be devised which will permit the restraining and periodic removal of oxide build-up in the hearth during continuous casting. One possible technique could utilize a water cooled dam and a skimming rake.

4.2.1.4 The main chamber must have, among its many considerations, easy access to all its parts, both internal and external. The nature of the operation is such that it is paramount for both health and safety considerations to be able to thoroughly clean and remove contamination products generated during the melt process. Melting of depleted uranium alloys results in distillation of the more radioactive daughter products from the natural decay of U₂₃₅. In addition, the operation produces some quantity of finely divided depleted uranium oxide. In keeping with these facts will be the necessity of a ventilation system such that when the chamber is opened, flow will be directed inward and negative pressure would then prevent the escape of contaminants into the work area. Therefore, the body of the chamber shall provide ports suitable for the attachment of a ventilation system.

4.2.1.5 It is desirable that view ports be located to permit observation of the melt and cast zones.

4.2.1.6 Once having cast the specific ingot, some method of cooling to a point where it may safely be removed should be addressed. An ingot at red heat will oxidize rapidly when exposed to air. This ability to cool will, of course, have a marked effect on cycle time and consequently on the throughput of the furnace.

4.2.1.7 All chambers of the furnace shall be equipped with an inlet and valve to permit backfill with inert gas.

5.0 QUOTATION REQUIREMENTS

5.1 Specification Compliance. The Vendor must address each item of this specification and describe how his equipment meets or exceeds the specification requirements. Vendor shall not limit his narrative to only the requirements of the specification but shall include any additional description of features which may enhance his equipment or point out advantages in his design or equipment. Descriptive literature describing the equipment quoted shall be incorporated with the quotation. The Vendor shall definitely state in his quotation whether the equipment complies or does not comply with every requirement of this specification and the Vendor shall state in detail where the offered equipment does not comply. The Vendor is free and is encouraged to recommend different or additional features that would increase the equipment productivity or reliability.

5.2 Pricing. Pricing must be firm for the equipment described under paragraph 1.0 and 4.0 and for the tooling needed to meet the requirements stated herein. Prices must be quoted on the equipment and tooling as would be required to meet the three production levels as stated in paragraph 3.3 and under the bases given in paragraphs 3.4 and 3.5. All prices shall be quoted in current prices and shall be firm for the specified time indicated in the Request for Proposal. Indication of future pricing trends may also be supplied for information and future consideration. The Vendor's terms and conditions shall be stated.

5.3 Delivery. A delivery schedule shall be submitted along with the quotation for each piece of equipment.

5.4 Operational Manuals. At a minimum of eight (8) weeks prior to delivery of equipment, the Vendor agrees to supply three (3) copies of the complete installation, operation and maintenance manuals.

5.5 Warranty. The Vendor shall state his standard warranty for the equipment to be supplied. Warranty shall commence upon final acceptance of the system at Nuclear Metals, Inc.

5.6 Tool Design. Tool design (if applicable) and quick change features (if applicable) must be briefly described for each item of tooling. An estimate of tool life shall be given. It is recognized that the depleted uranium-titanium alloy may not be familiar to the Vendor and that the tool life estimates can only be the Vendor's best judgement. Drawings of tooling must be submitted.

5.7 Installation Information. Installation of the equipment to the point of power supply shall be the responsibility of the Vendor, however, sufficient information must be furnished to Nuclear Metals, Inc. a minimum of twelve (12) weeks prior to shipment.

5.7.1 Layout drawings of the assembled equipment shall be furnished with the quotation.

5.7.2 Special Foundation(s) requirements shall be described in the proposal. The Vendor shall be required to supply drawings of any special foundation requirements. Nuclear Metals, Inc. shall construct the foundation(s) per the Vendor's drawing and specification.

5.7.3 Utility requirements for the equipment and its accessories shall be listed in both connected requirements and usage as follows:

a. Electrical Kw load, voltage and estimated usage of each major piece of equipment as well as total usage.

b. Number of motors, voltage and frame size of each.

c. Compressed air pressure required, air pipe sizes and usage in CFM.

d. Any other utility requirements not listed such as cooling water and special conditions as grounding, vibration or electrical isolation.

5.7.4 Vendor assistance in installation and start-up shall be specified.

5.7.4.1 All vendor personnel involved in installation will be required to attend a safety briefing.

5.7.4.2 Vendor's personnel will be supplied with safety glasses, safety shoes and uniforms for the duration of the installation and operational test.

5.8 Normal Preventative/Maintenance Requirements. Normal preventative/maintenance requirements that are considered routine must be listed along with the maintenance labor time and material requirements.

5.9 Spare Parts List. A list of recommended spare parts for the furnace and its accessories must be supplied along with the current price and delivery of each part. The Vendor shall specify those parts which are in normal inventory, and those parts which should be in inventory at Nuclear Metals, Inc.

5.10 Training and Technical Assistance. The Vendor shall furnish the required technical assistance during installation. The Vendor shall supply training of operating and maintenance personnel to insure safety, maximum performance and efficient and proper maintenance.

5.11 Production Rate. The guaranteed production rate (floor-to-floor time) of equipment shall be stated in the Vendor's proposal.

5.12 Auxiliary Support Equipment. The auxiliary support equipment to operate and maintain the equipment shall be listed. If such equipment is supplied by the Vendor, it shall be quoted in the same manner as the prime equipment. If such equipment is not supplied by the Vendor, the Vendor shall supply Nuclear Metals, Inc. with the necessary specification for purchase of such equipment.

5.13 Reliability and Maintainability. The Vendor shall provide Nuclear Metals, Inc. with data for prediction of near time for repairs and mean times to failure for his equipment.

5.14 Layaway Provisions. In the event that manufacture is delayed or ceases, Nuclear Metals, Inc. shall be responsible for putting the equipment in layaway condition. The Vendor shall provide Nuclear Metals, Inc. with the necessary procedures for putting equipment in layaway and maintaining equipment during layaway. The Vendor shall define all components which are subject to deterioration during layaway.

DATA SHEET

PHYSICAL CONSTANTS OF URANIUM

Density, gm/cm ³	19.13
Melting Point, °C	1133
Boiling Point, °C	3900
Heat of Fusion, KCal/Mole	2.5-3.0
Heat of Vaporization, KCal/Mole	93
Electrical Resistivity, 25°C u ohm-cm	2-4 X 10 ⁴

Specific Heat, Cal/(Mole) (°C)

27°C	6.649
774 ()°C	10.147
827°C	9.147

Enthalpy (H_T) Cal/Mole

827°C	10.430
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Entropy, Cal/(Mole) (°C)

827°C	24,761
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Thermal Expansion, 10⁻⁶ per °C

From X-ray Data,

Direction Parallel To Axis	<u>25°-125°C</u>	<u>25°-325°C</u>	<u>25°-650°C</u>
(100)	21.7	26.5	36.7
(010)	- 1.5	- 2.4	- 9.3
(001)	23.2	23.9	34.2
Volume	45.8	48.6	61.5

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